RESONANCES IN THE CAPTURE OF PROTONS BY SILICON

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Submitted in partial fulfillment of
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MASTER OF SCIENCE

United States Naval Postgraduate School
Monterey, California

1958

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This work is accepted as fulfilling
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ABSTRACT

Resonances in the capture of protons by the three natural occuring isotopes of silicon were investigated using a proton beam from the 2-Mev Van de Graaff generator at the U. S. Naval Postgraduate School. Thin targets of high purity natural silicon, separated Si^{29} isotope, and separated Si^{30} isotope were bombarded by protons within the energy range 0.85 to 2.03 Mev. The gamma-ray yield was observed as a function of the proton energy which was measured by means of a magnetic analyzer. Resonances from the $Si^{29}(p,\gamma)P^{30}$ reaction were found at proton energies of 920, 960, 1309, 1334, 1479, 1515, 1648, 1671, 1692, 1752, 1777 and 1857 kev. Resonances from the $Si^{30}(p,\gamma)P^{31}$ reaction were found at proton energies of 945, 989, 1108, 1178, 1188, 1214, 1221, 1263, 1297, 1303, 1307, 1329, 1353, 1397, 1406, 1425, 1491, 1498, 1519, 1526, 1606, 1667, 1675, 1701, 1777, 1811, 1814, 1821, 1836, 1882, 1897, 1924, 1944, 1977, 2009 and 2024 kev. There were no resonances found which resulted from the $Si^{28}(p,\gamma)P^{29}$ reaction.

The writers wish to express their appreciation for the assistance and encouragement given them by Professor Edmund A. Milne and Professor John N. Cooper in this investigation as well as to Kenneth C. Smith who provided assistance in the maintenance of electronic equipment.

This project was supported in part by the Office of Naval Research.

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1. Introduction

This investigation was conducted to determine the resonances in the Si(p,γ)P reaction using proton energies of from .85 to 2.03 Mev. and to assign these resonances to the particular silicon isotope responsible.

Naturally occurring silicon is composed of three isotopes as follows:

Isotope	Percentage (Wt.)
Si^{28}	92.21
Si^{29}	4.70
Si^{30}	3.09

It is therefore apparent that when natural silicon is bombarded with protons, three separate and distinct simple capture reactions can occur. They are:

Si^{28} (p,γ)P^{29}
Si^{29} (p,γ)P^{30}
Si^{30} (p,γ)P^{31}

Of the three phosphorous nuclides formed, P^{29} and P^{30} are radioactive, decaying by B^+ emission with half lives of 4.6 seconds and 2.5 minutes respectively, while P^{31} is stable.

In this investigation a thin target of high purity natural silicon was bombarded with protons and a differential excitation curve drawn. This curve included resonance peaks for all three of the above reactions. Separated isotopes of Si^{29} and Si^{30} were then similarly irradiated and curves drawn for each. It was then possible to attribute each resonance on the natural silicon curve to a specific reaction.

2. Resonance and Decay Phenomena.

Since 1930 various types of particle accelerators have been developed which have made it possible to study the excited states of light nuclei. When a nucleus is bombarded by a light particle (in this case, a proton) the particle may be merely scattered or a compound nucleus may be formed which can decay in a number of different ways. If decay is by gamma emission, the excitation function for the compound nucleus can be obtained by measurement of the gamma ray intensity as the incoming particle energy is varied. Resonances observed indicate discrete excited states of the compound nucleus and it is for this reason that the accurate determination of excitation curves is important. An extended analysis of the resonance phenomena has been given by Fowler, Lauritsen and Lauritsen.[1]

Examination of Table I indicates that when any of the three natural occuring isotopes of silicon is bombarded with protons of less than 2.47 Mev energy (laboratory system) gamma ray emission is the only decay mode energetically possible after proton capture.

Gamma rays may arise after inelastic scattering as well as after capture. The lowest energy levels of the three isotopes[2] under consideration are:

Si^{28}	1.78 Mev
Si^{29}	1.28 Mev
Si^{30}	2.24 Mev

It can be seen from the above tabulation that inelastic scattering resonances are not energetically possible for Si^{30} in the proton energy range 0.85 to 2.03 Mev. A proton of energy greater than 1.32 Mev in the laboratory system of coordinates striking a Si^{29} nucleus can cause inelastic scattering resonance but since the probability of this phenomenon is low (for both Si^{28} and Si^{29}) for the energies employed, it is assumed to add only insignificantly to observed resonances. Since no proton energies greater than 2.03 Mev were used in this investigation it is therefore assumed that in every event the excited phosphorus nucleus decays by the emission of quanta.

[1]W. A. Fowler, et. al., Rev. Mod. Phys., 20, 236 (1948)

[2]P. M. Endt and J. C. Kluyver, Rev. Mod. Phys., 26, 95 (1954)

3. Previous Investigations

The Si(p, γ)P reactions were first studied by Hole, Holtsmark and Tangen[3] in 1941 with proton energies between 300 and 550 kev. Later Tangen[4] conducted more detailed investigations in this same energy range and reported four sharp resonances which he attributed to specific silicon isotopes as follows:

Proton Energy (kev)	Isotope
326	Si^{29}
367	Si^{30}
414	Si^{29}
499	Si^{30}

The next reported investigations were made by Seiler[5] in 1955. His work with natural silicon verified the 414 and 500 kev resonances reported by Tangen and in addition indicated resonances at 622, 675, 698, 703, 732, 760, 778, 944 and 989 kev.

The next year, Milani, Cooper and Harris at Ohio State University[6,7] investigated the $Si^{29}(p,\gamma)P^{30}$ reaction using Si^{29} enriched to 80.8 percent. They reported resonances at 326, 414, 698, 731, 918 and 957 kev.

At about the same time, S. P. Tsytko and Iv. P. Antuf'ev[8], reported resonances obtained from natural silicon targets in the proton energy interval from 500 to 2600 kev. They reported new resonances at 619.5, 717, 753, 775, 800, 831, 895, 940, 980, 1520, 1618, 1635, 1647, 1663, 1680, 1699, 1774, 1810, 1849, 1879, 2520, 2543, 2553, 2557.5, 2570 and 2572 kev. They indicated that identification of the reaction corresponding to the resonances were made below 1000 kev. They found no resonances from the $Si^{28}(p,\gamma)P^{29}$ reaction among those identified. Identification was accomplished by the yield of

[3] N. Hole, et. al., Phys. 118, 48 (1941)

[4] R. Tangen, Kgl. Norske Vid. Selsk. Skr. NRI (1946)

[5] M. R. Seiler, M. S. Thesis, Ohio State University 1955 & Phys. Rev. 99, 430 (1955)

[6] J. N. Cooper, Annual Report by the Ohio State University Research Foundation, March 16, 1955 - March 15, 1956. RF Project 440, Report No. 6.

[7] S. Milani, et. al., Phys. Rev. 99, 645 (1955)

[8] S. P. Tsytko and Iv. P. Antuf'ev, J. Exptl. Theoret Phys. (U.S.S.R.) 30, 1171 (June, 1956).

positron activity from thin natural silicon targets.

Very recently, Seagondollar, Woods, De Sousa and Glass[9] have bombarded hyperpure natural silicon at the University of Kansas with protons of energies between 300 and 1400 kev. They report resonances at 326, 369, 414, 501, 622, 697, 776, 836, 943, 957, 979, 1202, 1205, 1291, 1327 and 1394 kev.

Table II summarizes the resonances reported to date in the energy range covered in the present investigation (i.e. 850 to 2030 kev). Where a resonance has been identified with a particular reaction, this fact is indicated.

4. Equipment

Protons used in this investigation were produced in the 2-Mev Van de Graaff electrostatic accelerator at the U. S. Naval Postgraduate School. This horizontal accelerator was manufactured by High Voltage Engineering Cooporation. The proton source is similar to the radio frequency ion source described by C. D. Moak[10]. The accelerated protons were passed through a 25 degree magnetic analyzer to separate ions of unwanted mass. The current through the coils of the magnet was measured by a Leeds and Northrup potentiometer. The energy width of the proton beam was defined by passing the beam through a slit 0.8 millimeters wide at a distance of 2.1 meters from the center of the magnet.

The target chamber was constructed so that the target could be insulated from the chamber and at the same time be positioned as far as possible inside a well type scintillation crystal. To accomplish this, a 7.8 centimeter long glass cap (¾" O.D.) was fitted on the end of the metal target chamber using a high vacuum metal to glass coupling. (See Fig. 1 and 2.) A silver wire insulated with teflon was brought into the metal chamber using a Kovar-glass seal and led through the chamber to the target holder. Silver target holders were constructed so as to fit onto the end of the silver wire and provide a firm holder for the targets. With this arrangement it was possible to connect the

[9] L. W. Seagondollar, et al., Bulletin Am. Phys. Soc. Ser. II, Vol. 2, No. 6, 304 (1957)

[10] C. D. Moak, et al., Nucleonics, 9, No. 3, 18 (1951)

target electrically to a current integrating circuit. A silver wire loop was placed around the beam just ahead of the target and the wire run out of the chamber through another Kovar-glass seal and through a 10 megohm resistor to the negative terminal of a 300 volt battery. This provided a Faraday Cage arrangement which repelled from the Faraday Cage any negative ions formed in front of the Faraday Cage by ionization of residual gas molecules and also prevented the escape of any electrons from the target.

A relay of the integrating circuit was connected to the count switch of a decimal scaler and to a timer. Similarly connected was a solenoid-operated beam shutter which cut the proton beam off and on. This made it possible to commence and stop target bombardment, current integration, timing and counting — all simultaneously. Use of the beam shutter automatically prevented overheating and unwanted bombardment of the target without disturbing the proton beam. Also a simple on-off switch between the shutter coil and the integrator permitted bombardment of the target at any time while the integrator was not running. This simplified focusing of the proton beam.

An auxiliary oil diffusion and fore pump system was installed between the slits and the target. Valves were so arranged so as to make it possible to isolate the target chamber when a change of targets was necessary. The total vacuum system was divided into three parts — the main system ahead of the slits, the auxiliary system and the target chamber. Such an arrangement made it possible to change targets and restore a vacuum in less than three minutes. The target chamber was roughed out before reopening to the auxiliary system. An ionization gauge tube in the auxiliary system made it possible to insure a high vacuum in this system before reopening it to the main system. (See Fig. 3.)

The gamma ray yield from the target was measured by a two-inch diameter, well-type, thallium-activated sodium iodide crystal mounted on a Dumont 6292 photomultiplier tube. The signals were amplified and counted by the scaler previously mentioned. A photograph and block diagram of the detection equipment is presented in Fig. 3 and 4 respectively.

5. Target Production

All silicon and LiF targets were made by evaporating "in vacuo" on to clean tantalum backing. The target backings were made by cutting discs one centimeter in diameter from 5 mil tantalum sheet. A considerable amount of difficulty was experienced in making satisfactory silicon targets until a tungsten sheet "boat" was used to hold the silicon. Prior to using tungsten, "boats" made of beryllium oxide, tantulum and carbon were all used unsuccessfully. The tantulum could not be brought to the high temperature necessary to coat the silicon oxides and the beryllium and carbon excessively contaminated the targets.

Thin natural silicon targets were first made using both the monoxide and dioxide form. Later hyper-pure silicon was used (99.9% pure natural silicon) and not only were the targets easier to make but the resonances obtained were more sharply defined on a decreased background.

Electro-magnetically separated isotopes of Si^{29} and Si^{30} were obtained from the Oak Ridge National Laboratories. The Si^{29} was enriched to 83% and contained 16% Si^{28} and less than 1.0% Si^{30}. The Si^{30} was enriched to 72.56% and contained 25.9% Si^{28} and 1.56% Si^{29}. Both were in the SiO_2 form. Thin targets were made with each of these enriched isotopes. In each case evaporations were made until smooth targets were obtained with thicknesses of about one kev at 1.5 Mev proton beam energy. Observation of the color changes and uniformity of target color during the evaporation process aided in estimating target thickness prior to proton bombardment.

6. Experimental Procedure

During all operations of the accelerator one man was stationed at the console to operate the accelerator and the magnetic analyzer. Prior to each operation the magnet was slowed cycled twice to minimize the hysteresis effect and once operation was commenced the magnetic current was always increased so as to stay on the same hysteresis loop. At each new setting the potentiometer was checked against a standard cell before recording the magnet current.

A second man was stationed near the target chamber and operated the current integrator and detection system. An intercom between the two stations allowed the second man to assist the first in controlling the machine by transmitting readings of beam current and corona stabalizer balance meters. Data was recorded by the man near the target chamber.

Prior to investigating silicon, the machine was calibrated using the LiF targets. The prominent fluorine resonances[11] as well as the $Li^7(p,n)Be^7$ threshold were used to construct the calibration curve presented on Fig. 5. Before bombarding the silicon targets the counting circuit was carefully biased to exclude all signals less than 1.12 Mev. This was accomplished by adjusting the bias on the linear amplifier using a Zn^{65} source. This insured that none of the annihilation quanta resulting from the P^{29} and P^{30} β^+ emission reached the scaler.

Preliminary runs were made from 870 kev to about 2000 kev with each silicon target, taking steps of about 0.001 amperes corresponding to energy steps of 1.64 kev. Rough plots were made and in subsequent runs the steps were reduced to approximately 500 kev while traversing resonances. At each magnet setting the number of counts, the magnet current and the time of observation was recorded. This time of observation at each point varied from about five seconds in the preliminary runs to about 30 seconds in the final runs.

Three excitation curves were drawn (natural silicon, Si^{29} and Si^{30}) after several traverses through the complete energy range. Calibration was checked each time the machine was operated. The calibration sometimes shifted as much as 10 kev from day to day.

[11] S. E. Hunt and K. Firth, Phys. Rev. 99, 786 (1955)

Upon completion of the three final excitation curves the energy values of the more prominent peaks on each curve were rechecked by a quick change of targets while traversing between the peak and a nearby fluorine resonance or vice versa.

Lastly, a determination was made of both time-dependent and time-independent background radiation. This was done by traversing the complete range in intervals of 10 kev, with a blank target which had been made and handled in the precise manner as the other targets except it had been vacuum coated from a clean tungsten "boat". Readings were taken at each energy step using a short and long observation time (about five and 30 seconds). In addition, a reading was taken for 15 seconds while the beam was intercepted by the beam chopper about three feet ahead of the target. This latter determination enabled the time-dependent background due to cosmic rays and Bremsstrahlung radiation to be obtained as a function of proton energy.

7. Resolution of Equipment

The manufacturer of the Van de Graaff and magnetic analyzer claim a proton energy resolution of about 0.2 percent at one Mev. This would produce an energy width of two kev at one Mev. Repeated determinations of resonance peaks during this investigation indicate that the resolution is at least this good. With the slit opening of 0.8 mm at a distance of 2.1 meters from the center of the magnet, several resonance peaks from the Si^{30} were measured with experimental widths of as little as 2.2 kev. It seems reasonable to attribute at least one kev of this width to target thickness. Assuming the natural width of these resonances to be negligible it appears that the proton beam energy width is two kev or slightly less. Considering the widths to add as the square root of the sum of their squares, a target width of one kev and a beam width of two kev gives a total width of 2.236 kev.

The stability of the magnetic analyzer current was generally excellent. A drift of less than 0.0001 amperes during the interval of bombardment was generally observed but at times the drift was as much as 0.0005 amps or 0.82 kev. It was noted that after a traverse over an energy range of about 500 kev (taking data in one kev steps)

the calibration would be in error sometimes as much as 2 kev. This error was attributed to current drift affecting the hysteresis and was minimized by recalibration each time primary calibration points were passed. The primary calibration points being the six fluorine resonances at 874.5, 935.1, 1283, 1348, 1375 and 1694 kev and the lithium neutron threshold at 1881 kev.

In view of the above discussion it does not appear unreasonable to assign a standard deviation of two kev to the results.

8. Results

The differential excitation curves obtained in this investigation are presented in Figures 6 through 23. Fig. 24 is a composite presentation of the three complete curves before background was subtracted. This figure was included to facilitate comparison of the curves although it is somewhat qualitative. It should be noted that the peaks in the natural silicon curve on this figure are arbitrarily doubled in height.

Tables III and IV list the resonances obtained from the reactions $Si^{29}(p,\gamma)P^{30}$ and $Si^{30}(p,\gamma)P^{31}$. All the resonances from the natural silicon were accounted for by these two reactions. This indicates that there were no resonances resulting from the $Si^{28}(p,\gamma)P^{29}$ reaction. This appears reasonable considering the stability of the Si^{28} nucleus and the resulting low excitation of the P^{29}.

Below 1000 kev the agreement with previously published data is generally very good. However, no indication whatsoever was found of the 895 kev resonance attributed to Si^{30}. The two resonances listed on Table II as 940 and 944 kev with the 940 attributed to Si^{30} are believed to be the same resonance. The present results indicate a strong Si^{30} resonance at 945 kev with a relative yield of 600. Above 1000 kev a total of sixteen resonances have previously been reported. Of the five resonances reported by Seagondollar, et al., only three were observed; the two resonances reported at 1202 and 1205 kev being unobserved in the present investigation. Of the eleven resonances reported by Tsytko and Antuf'ev, only eight were in agreement ($\pm$ 4 kev) with the present results.

As indicated by the excitation curves, flourine contamination

of the targets was never completely eliminated. Some carbon contamination was also present as indicated by the broad C^{12} resonance at about 1690 kev[12] which added to the background in all runs. The contamination present varied as different targets were used but as the technique of target making improved the amount of contamination steadily decreased.

Figures 25 and 26 are energy level diagrams for P^{30} and P^{31} which include the new levels determined in the present investigation.

It should be emphasized that the accuracy of the results obtained here depend on the accuracy of the primary calibration points used to construct the calibration curve.

[12]J. D. Seagrave, Phys. Rev., 85, 197 (1952)

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3. N. Hole, J. Holtsmark, and R. Tangen, Z. F. Phys., 181, 48 (1941).

4. R. Tanzen, Kgl. Norske Videnskab. Selskabs, Skrifter, NR 1 (1946).

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7. S. Milani, J. N. Cooper, and J. C. Harris, Phys. Rev. 99, 645 (1955).

8. S. P. Tsytko and Iv. P. Antuf', J. Exptl. Theoret. Phys. (U.S.S.R.) 30, 1171 (June, 1956).

9. L. W. Seagondollar, J. A. Woods, H. G. DeSouza and W. A. Glass, Bulletin of the Am. Phys. Soc. Ser. II, Vol. 2, No. 6, 304 (1957).

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11. S. E. Hunt and K. Firth, Phsy. Rev., 99, 786 (1955).

12. J. D. Seagrave, Phys. Rev., 85, 197 (1952).

13. C. Broude, L. L. Green, J. J. Singh, and J. C. Willmott, Phys. Rev., 101, 1052 (1956).

Silicon - Proton Reactions

Reaction	Q (Mev)	Bibliography Reference
$Si^{28}\ (p,\gamma)P^{29}$	2.724	2
$Si^{29}\ (p,\gamma)P^{30}$	5.5	2
$Si^{30}\ (p,\gamma)P^{31}$	7.290	2
$Si^{28}\ (p,n)P^{29}$	-14.9	13
$Si^{29}\ (p,n)P^{30}$	- 5.749	2
$Si^{30}\ (p,n)P^{31}$	- 5.1	2
$Si^{28}\ (p,d)P^{29}$	-14.976	5
$Si^{29}\ (p,d)P^{30}$	- 6.246	13
$Si^{30}\ (p,d)P^{31}$	- 8.388	13
$Si^{28}\ (p,\alpha)P^{29}$	- 7.70	2
$Si^{29}\ (p,\alpha)P^{30}$	- 4.6	2
$Si^{30}\ (p,\alpha)P^{31}$	- 2.389	2

Table I

Silicon Resonances from Previous Investigations
in the Range 850 to 2030 Kev.

Proton Energy	Reaction	Bibliography Reference
895	$Si^{30}(p,\gamma)P^{31}$	8
918	$Si^{29}(p,\gamma)P^{30}$	7
940	$Si^{30}(p,\gamma)P^{31}$	8
944		5,9
957	$Si^{29}(p,\gamma)P^{30}$	7,9
980	$Si^{30}(p,\gamma)P^{31}$	8,9
989		5
1202		9
1205		9
1291		9
1327		9
1394		9
1520		8
1618		8
1635		8
1647		8
1663		8
1680		8
1699		8
1774		8
1810		8
1849		8
1879		8

Table II

Resonances in the Capture of Protons by Si^{29}

Energy (kev)	Relative Yield	Experimental Half Width
920 ± 2	150	5.0
960 ± 2	70	6.5
1309 ± 2	470	5.4
1334 ± 2	800	6.9
1479 ± 2	300	5.0
1515 ± 2	1380	5.0
1648 ± 2	300	8.3
1671 ± 2	390	7.3
1692 ± 2	1300	6.0
1752 ± 2	1750	4.5
1777 ± 2	650	6.2
1857 ± 2	220	4.4

Table III

Resonances in the Capture of Protons by Si^{30}

Energy (kev)	Relative Yield	Experimental Half Width
945 ± 2	600	4.0
989 ± 2	680	3.3
1108 ± 2	150	4.1
1178 ± 2	600	4.8
1188 ± 2	200	3.5
1214 ± 2	1180	3.2
1221 ± 2	500	4.3
1263 ± 2	480	4.1
1297 ± 2	270	3.3
1303 ± 2	420	3.2
1307 ± 2	500	4.5
1329 ± 2	1490	4.3
1353 ± 2	270	3.8
*1379 ± 2	220 ± 50	3.0 ± 1.0
1397 ± 2	1720	2.5
1406 ± 2	1600	2.1
1425 ± 2	240	2.7
1491 ± 2	1850	3.1
1498 ± 2	1100	2.4
1519 ± 2	1400	2.6
1526 ± 2	200	3.7
1606 ± 2	280	2.1
1667 ± 2	270	2.5
1675 ± 2	380	2.3
1701 ± 2	790	2.9
1777 ± 2	1200	3.0
1811 ± 2	800	4.0
1814 ± 2	820	4.5
1821 ± 2	700	2.8
1836 ± 2	1350	3.5
1882 ± 2	2100	2.2
1897 ± 2	600	5.6
1924 ± 2	530	4.9
1944 ± 2	350	5.0
1977 ± 2	520	5.8
2009 ± 2	680	2.9
2024 ± 2	570	3.7

* Indication only.

Table IV

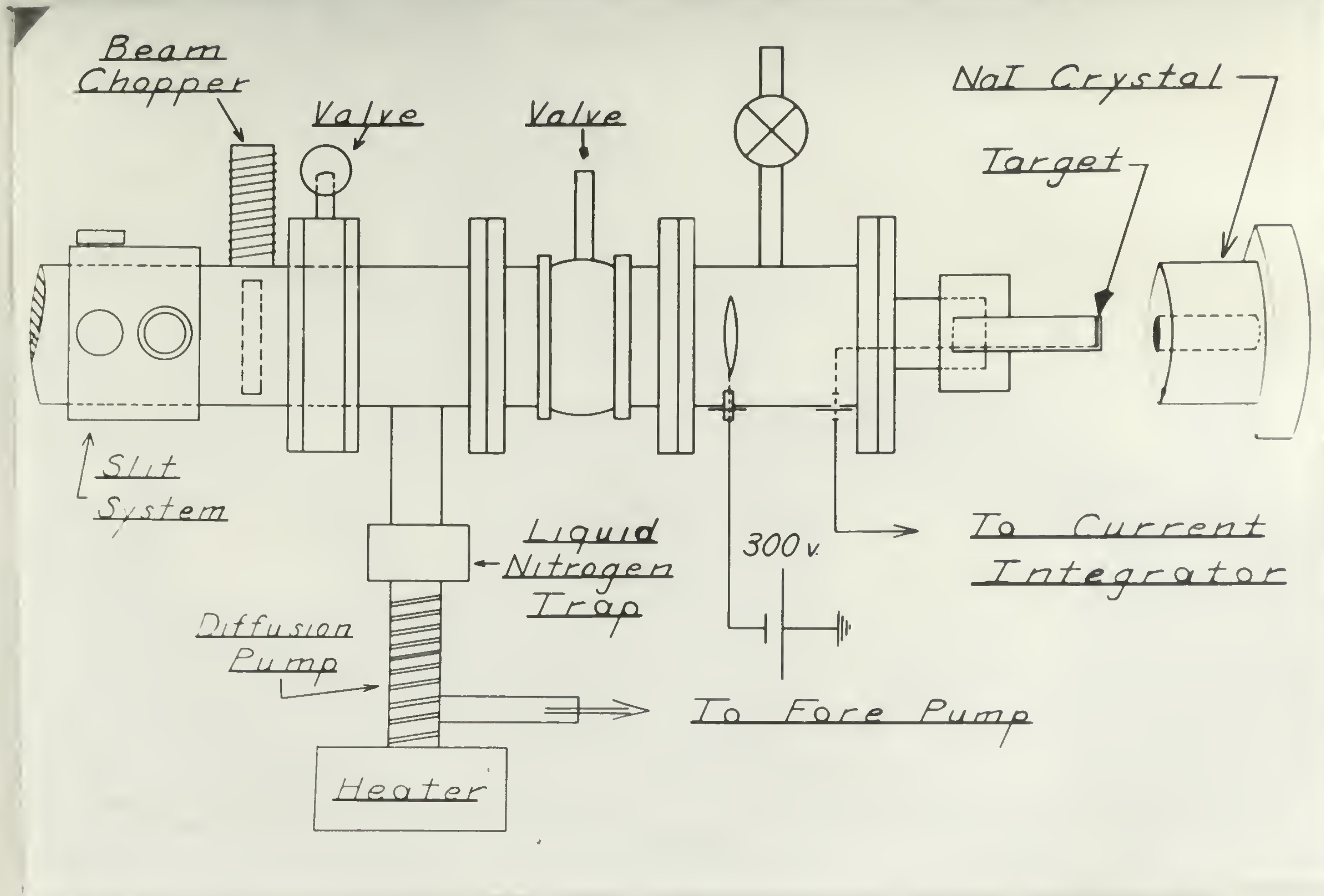


Figure 1

Schematic Diagram of Target Chamber

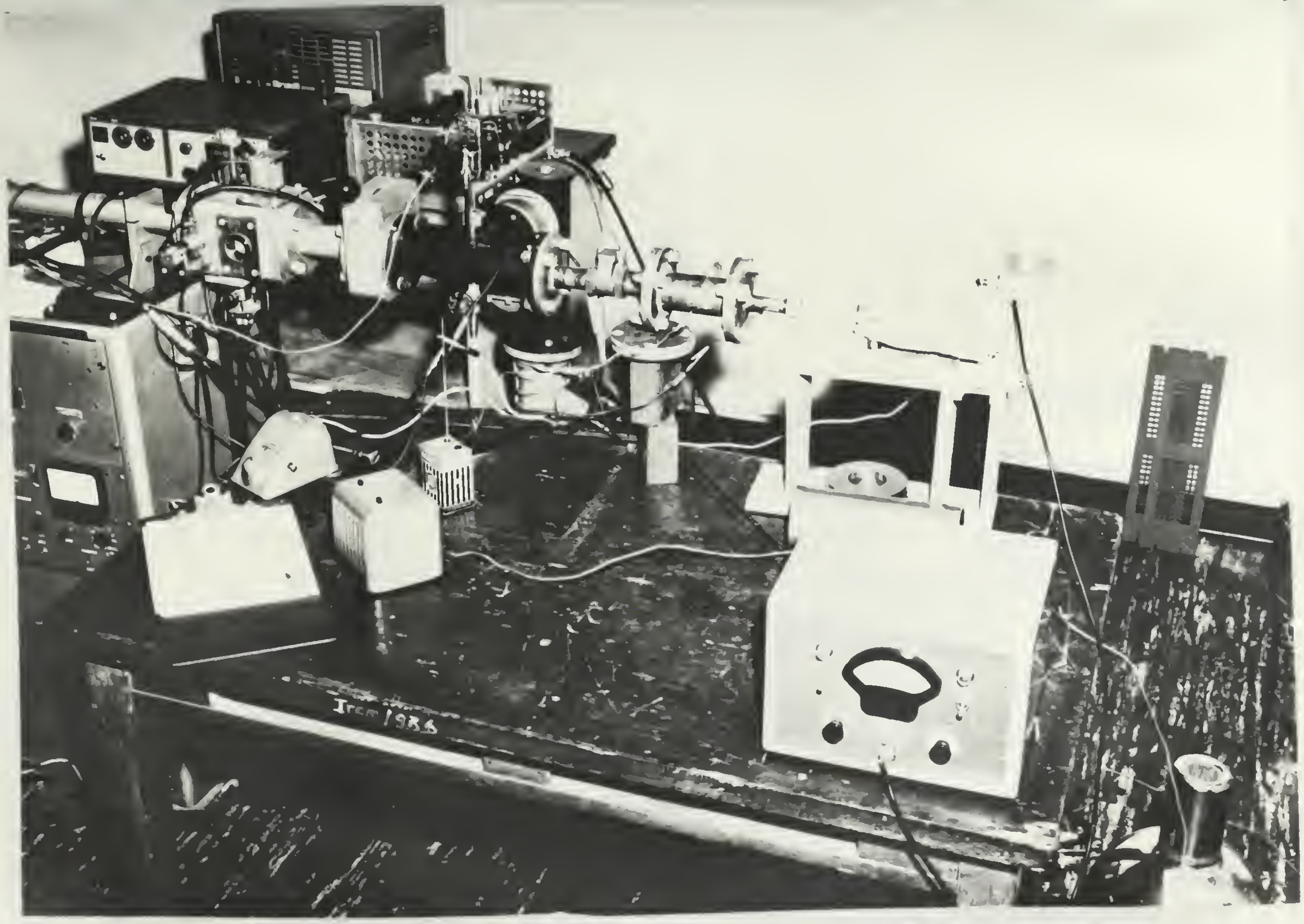


Figure 2

Photogragh of Target Chamber

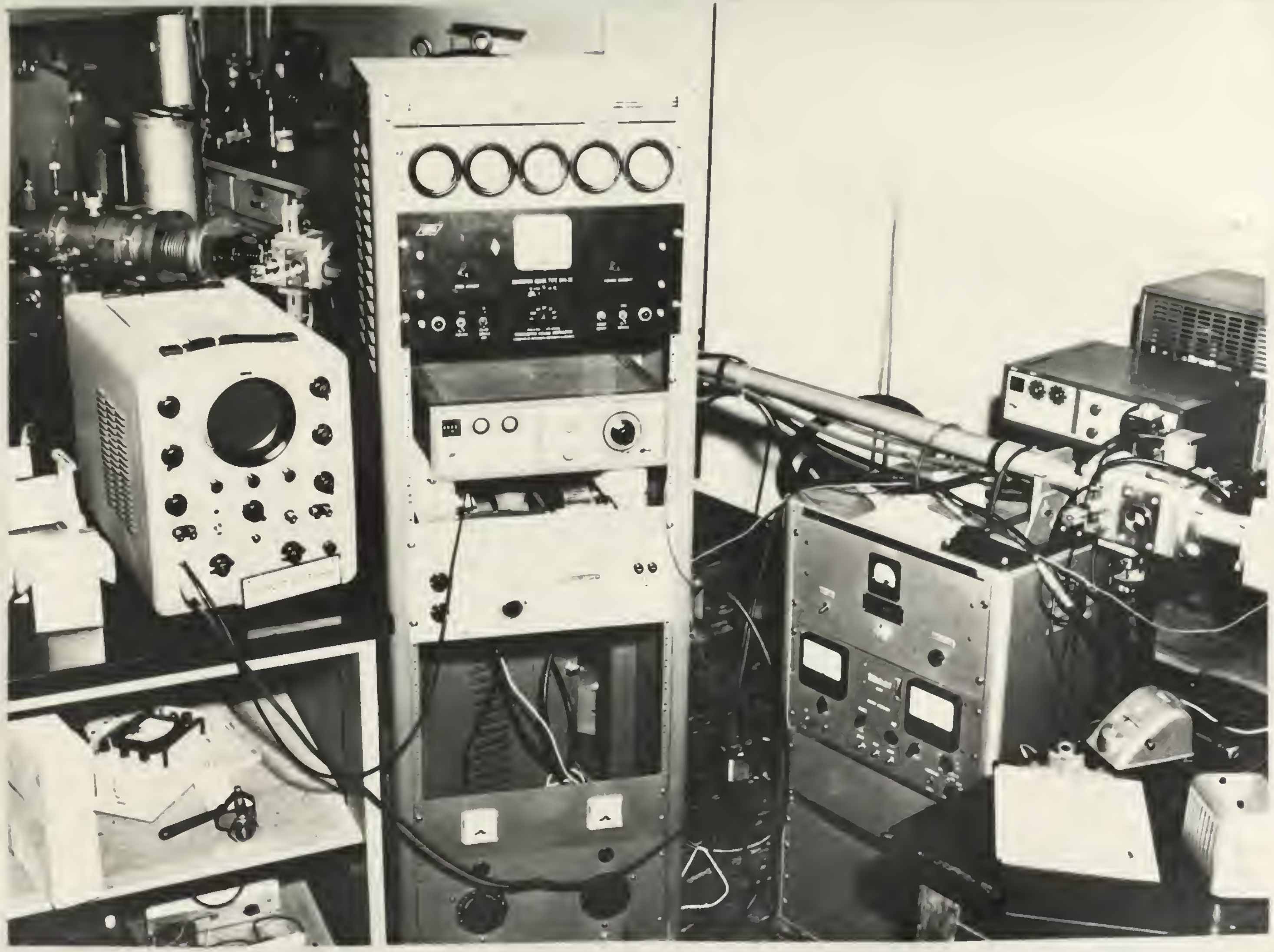


Figure 3

Photogragh of Associated Electronic Equipment.

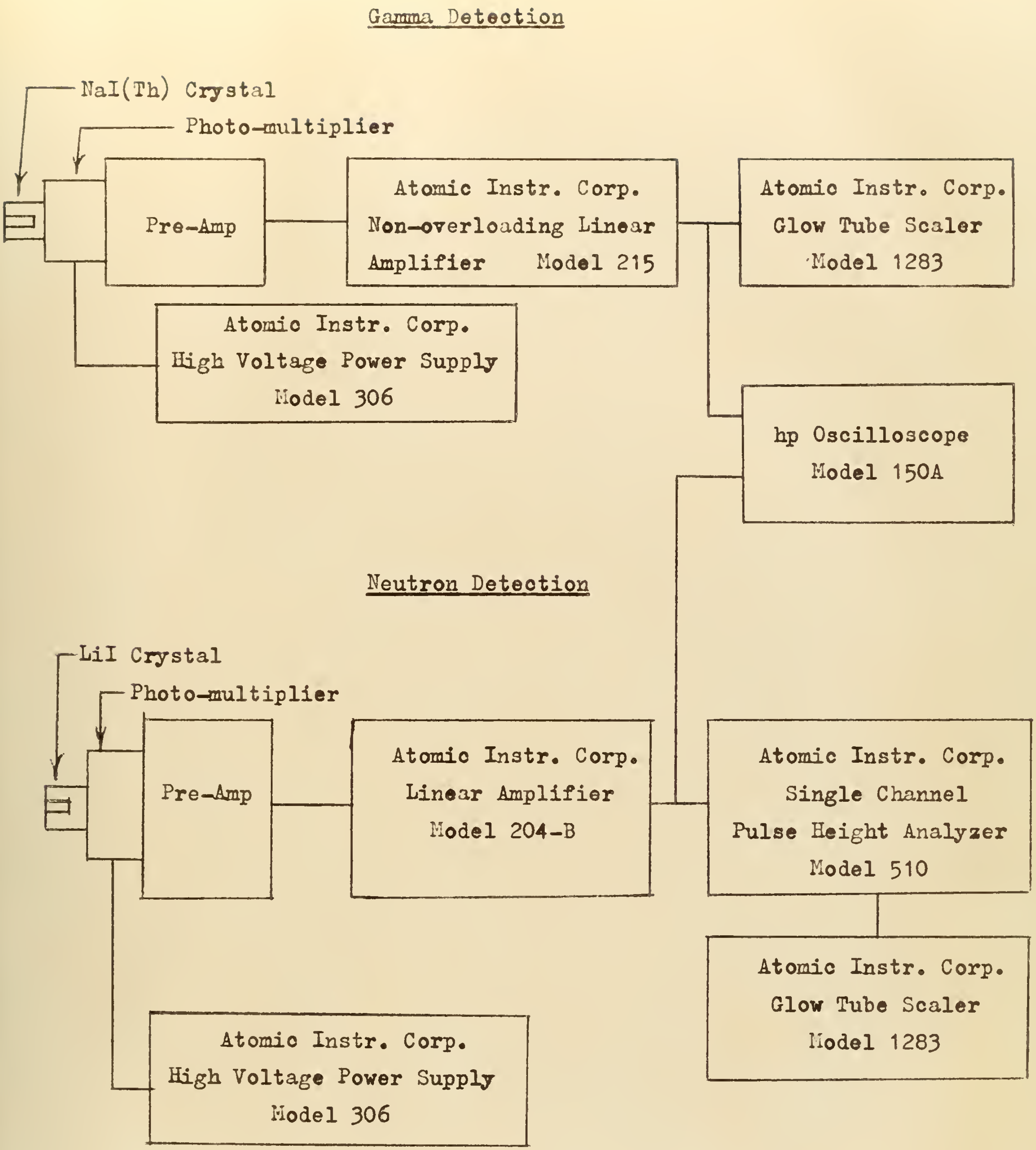


Figure 4

Block Diagram of Detection Equipment

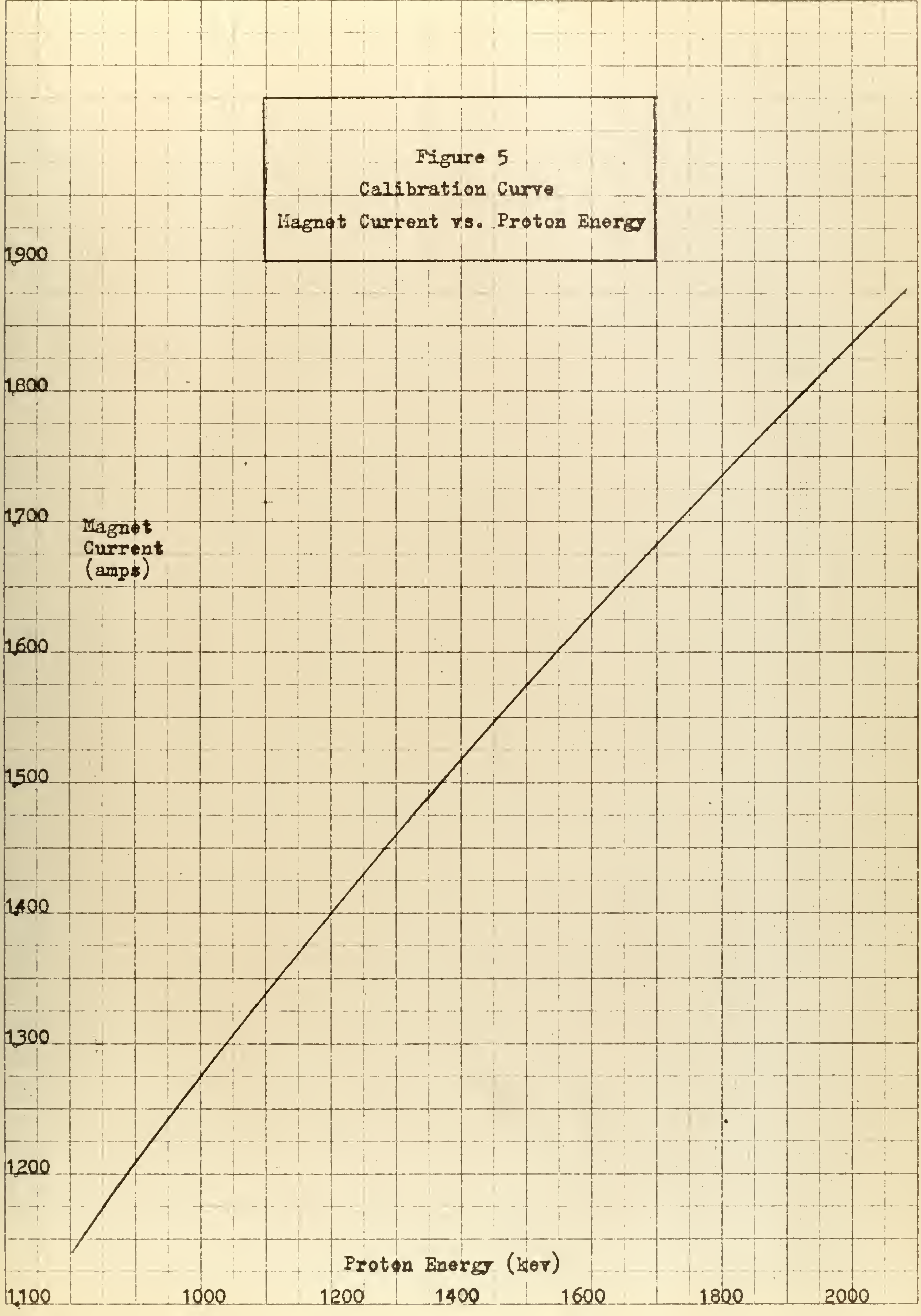


Figure 5
Calibration Curve
Magnet Current vs. Proton Energy

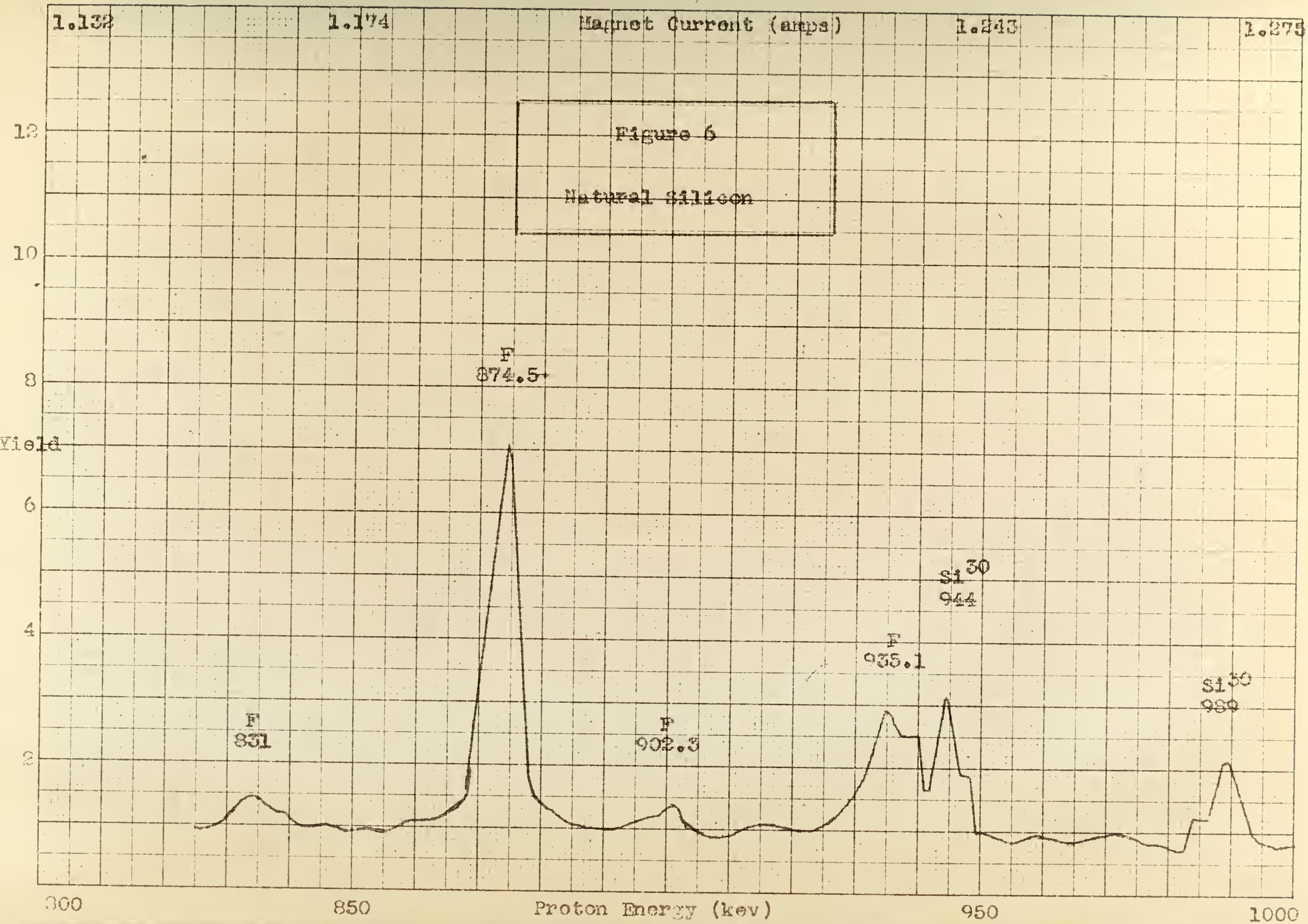
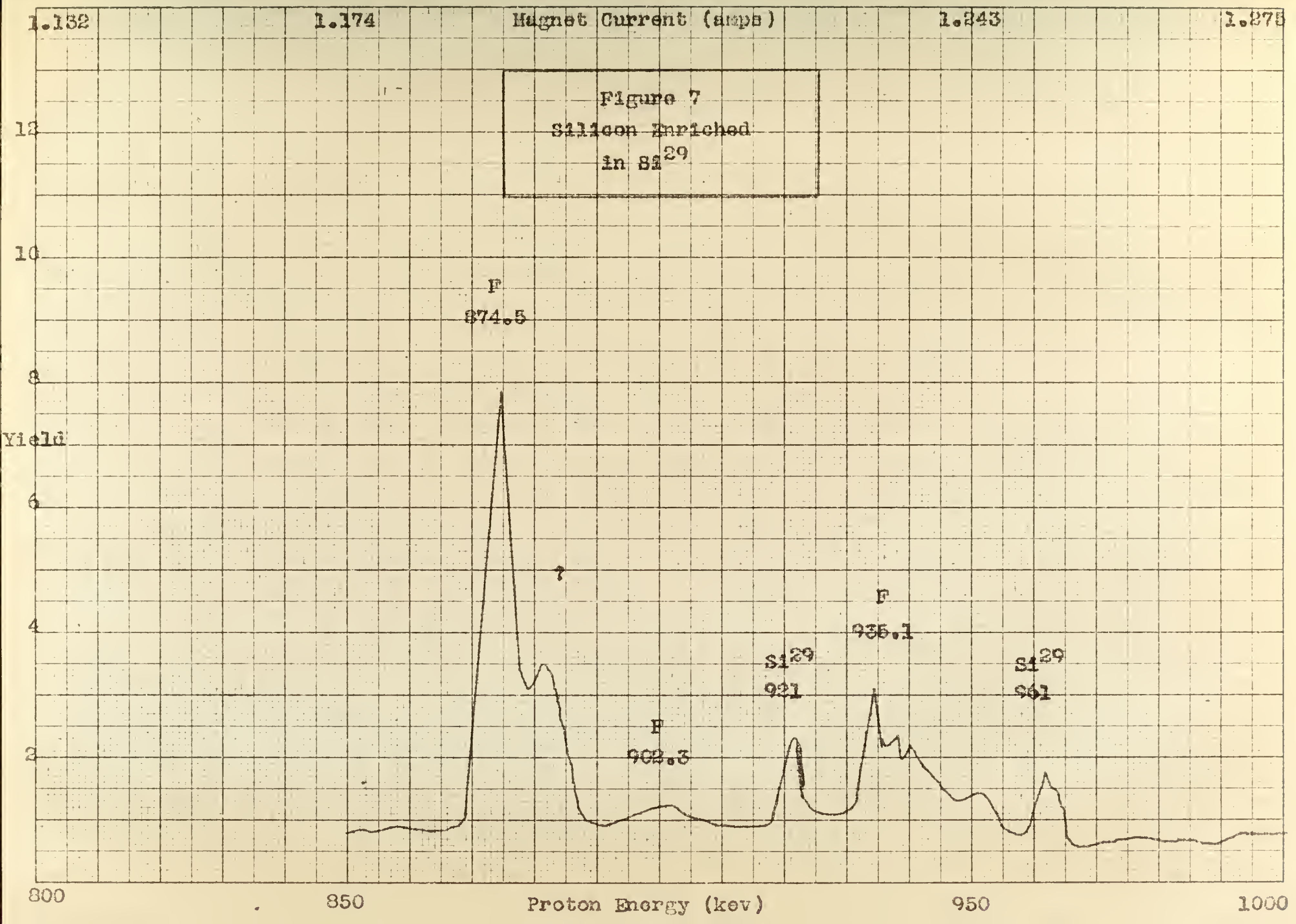


Figure 6

Natural Silicon



1.132
1.174
Magnet Current (amps)
1.243
1.275
Figure 7
Silicon Enriched
in Si29
12
10
8
Yield
6
4
2
F
874.5
?
F
902.3
Si29
921
F
935.1
Si29
961
800
850
Proton Energy (kev)
950
1000

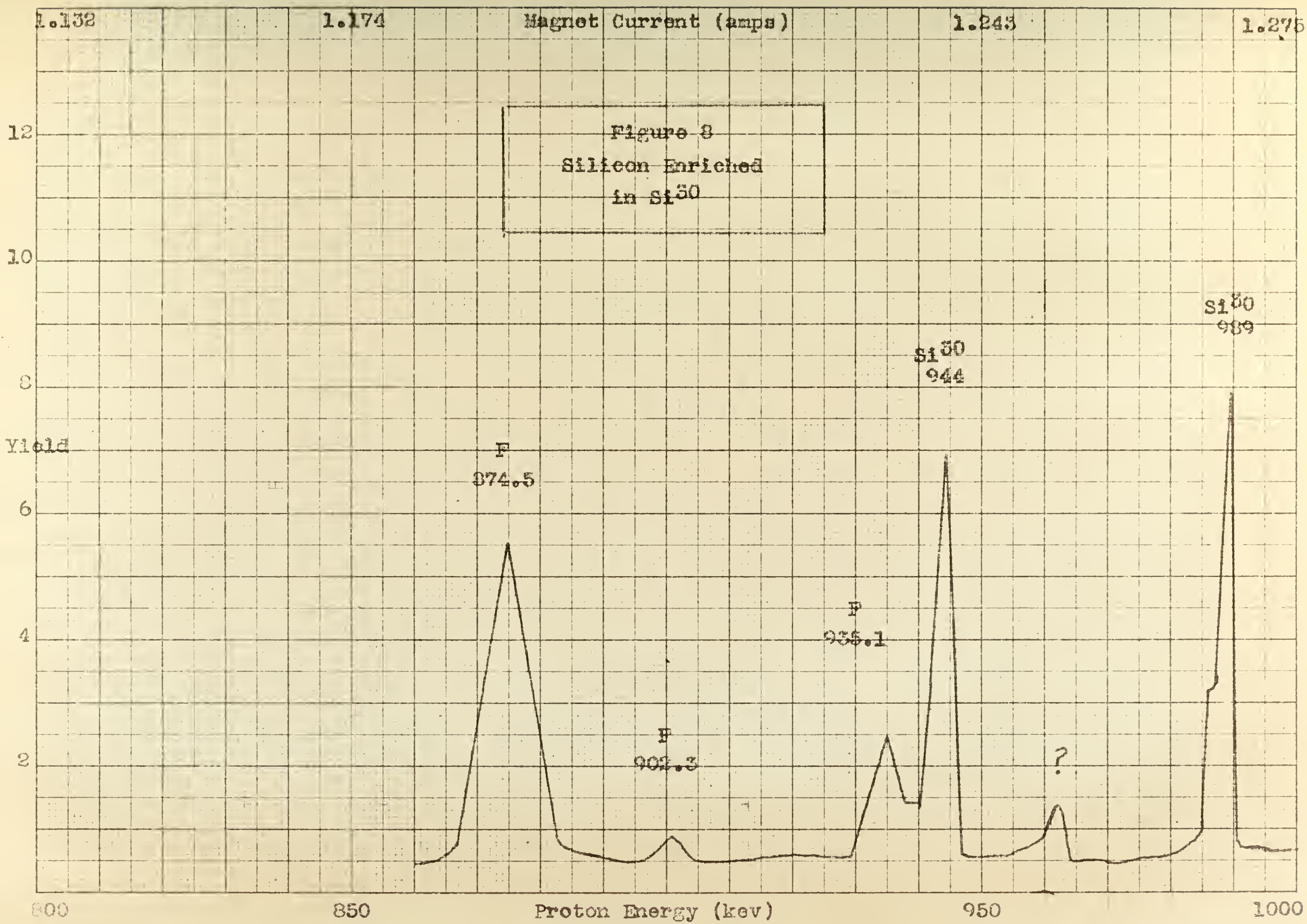
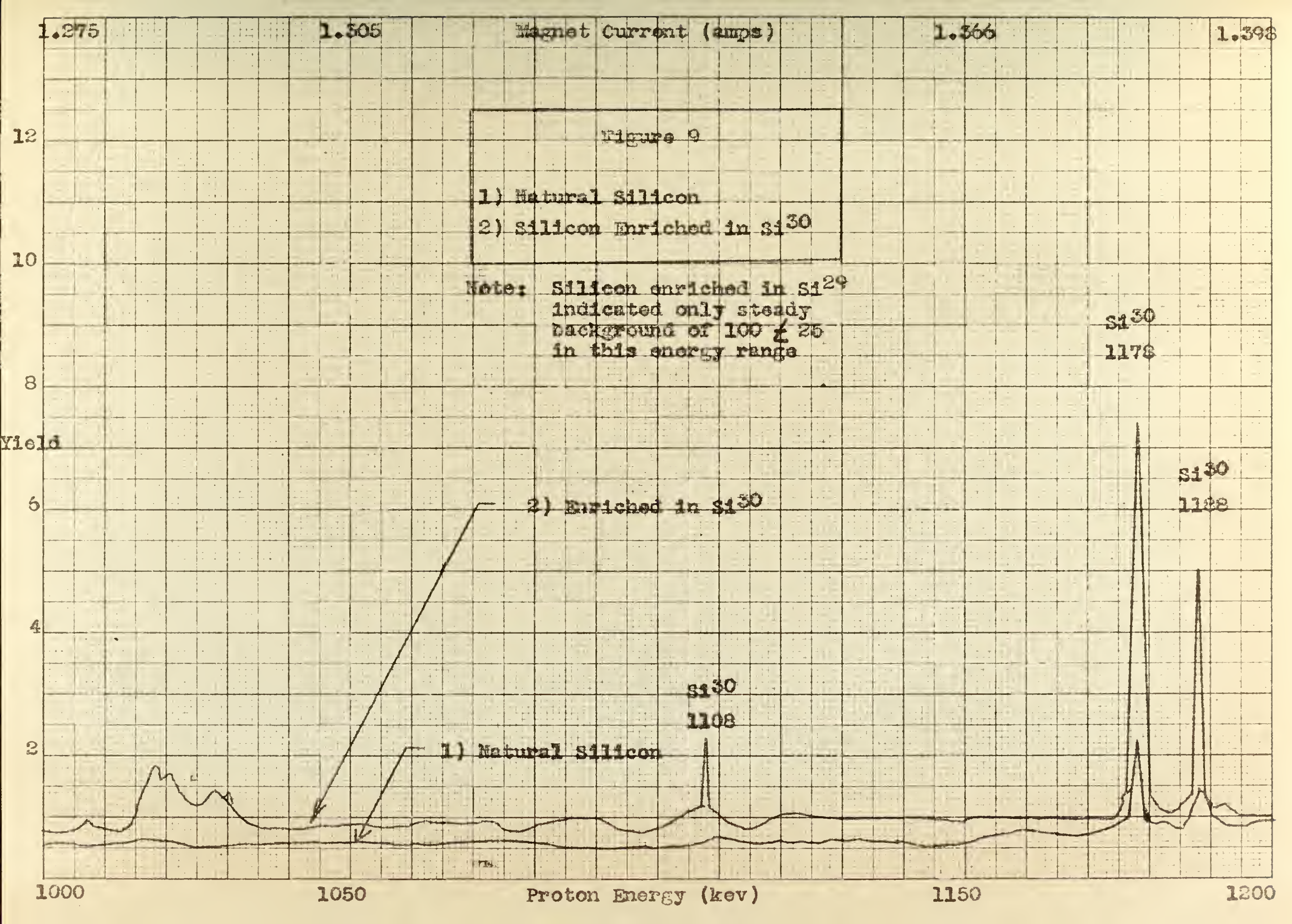


Figure 8
Silicon Enriched
in Si^{30}


1.275
1.305
Magnet Current (amps)
1.366
1.398
Figure 9
1) Natural Silicon
2) Silicon Enriched in Si^{30}
Note: Silicon enriched in Si^{29} indicated only steady background of 100 ± 25 in this energy range
Si^{30} 1178
Si^{30} 1188
2) Enriched in Si^{30}
Si^{30} 1108
1) Natural Silicon
Yield
12
10
8
6
4
2
1000
1050
1150
1200
Proton Energy (kev)

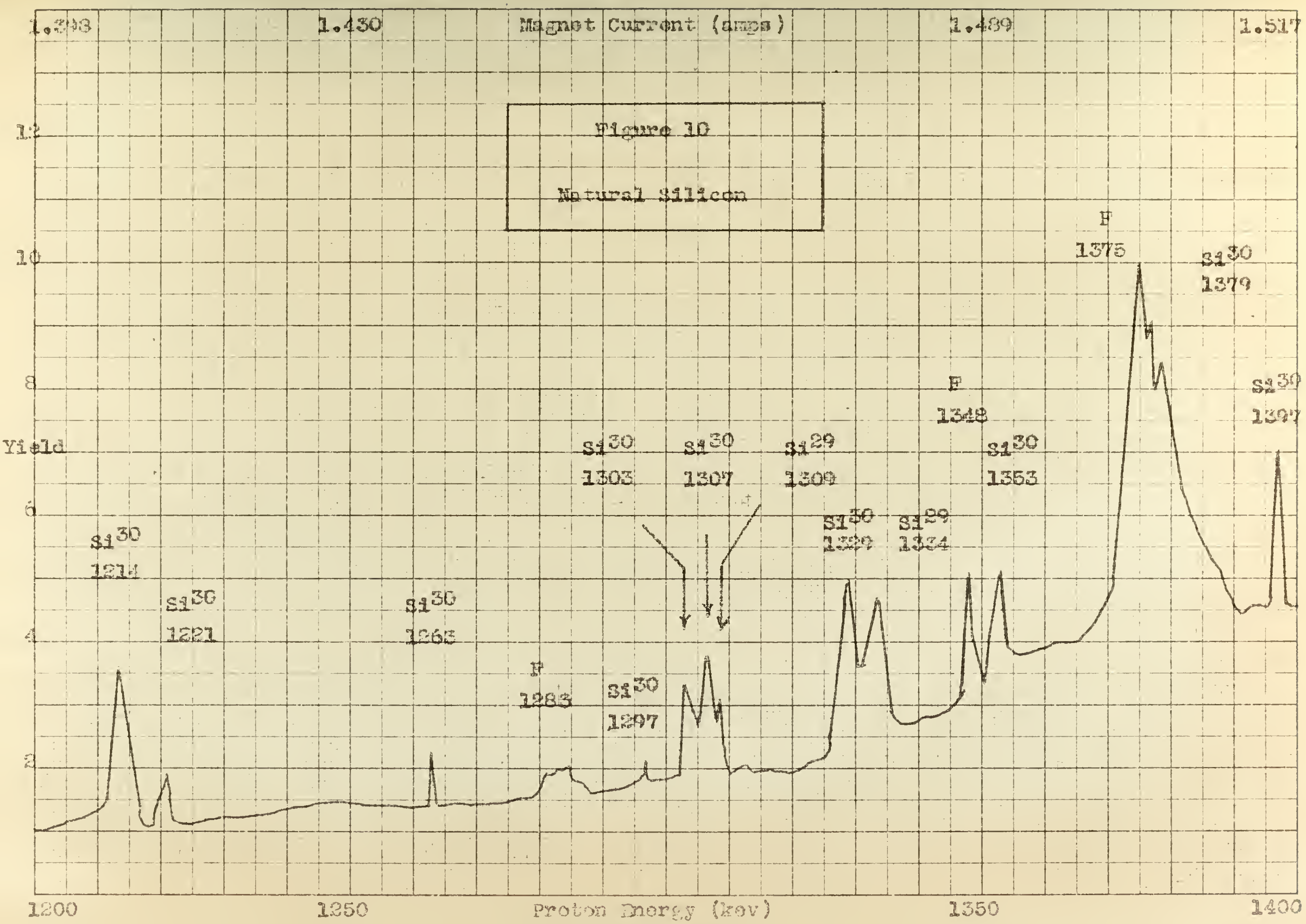


Figure 10

Natural Silicon

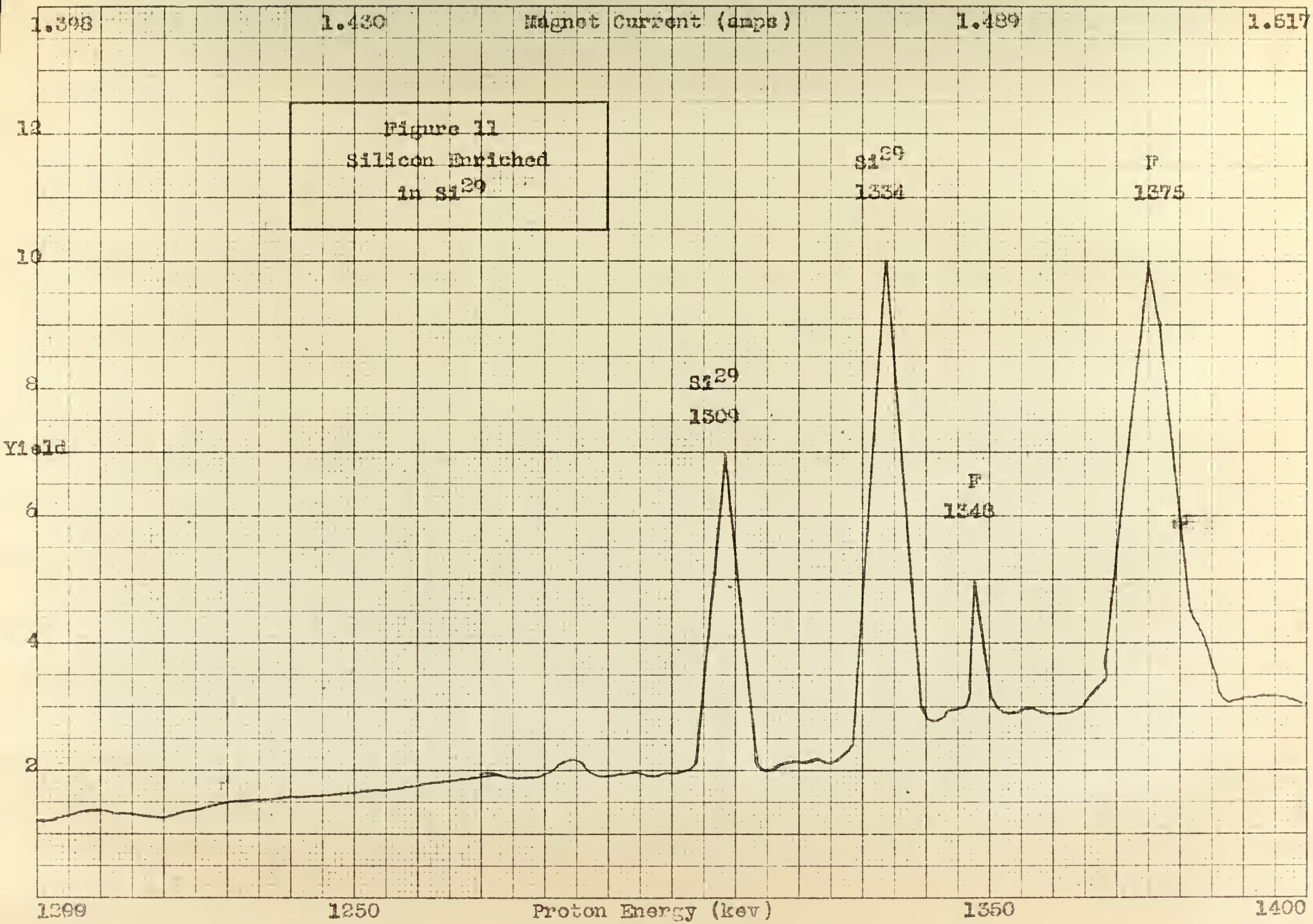


Figure 11
Silicon Enriched
in Si^{29}

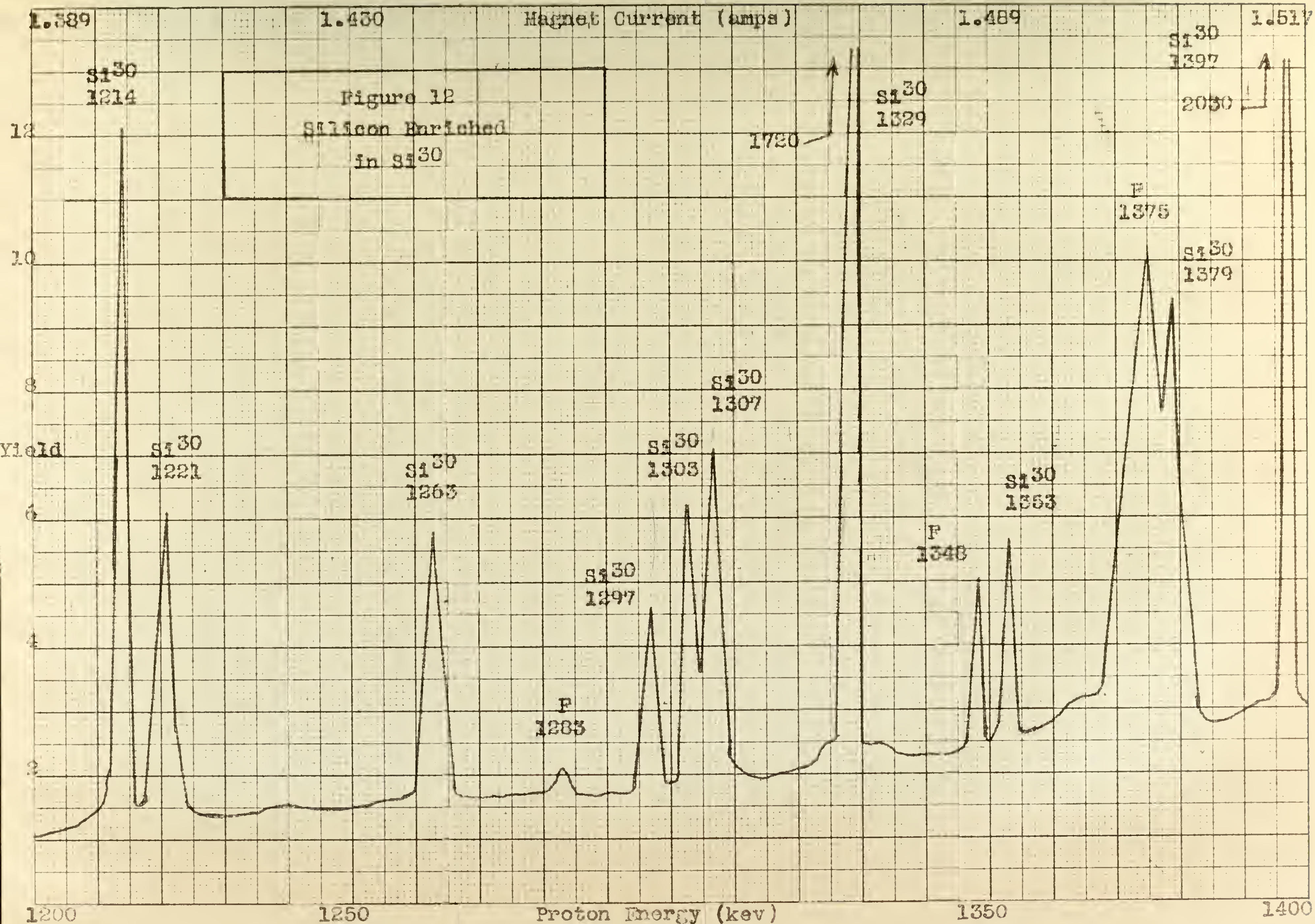


Figure 12
Silicon Enriched
in Si^{30}

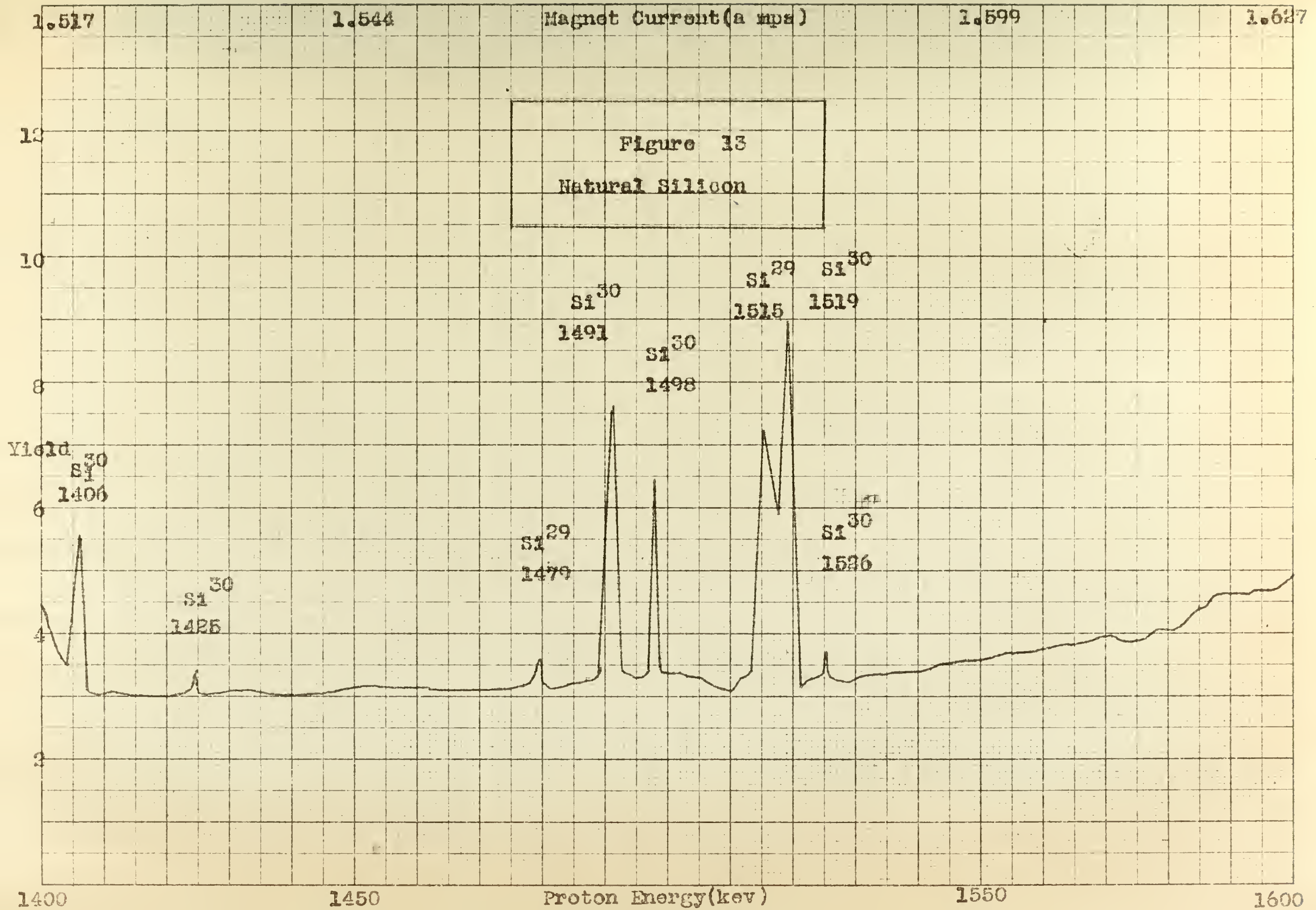


Figure 13

Natural Silicon

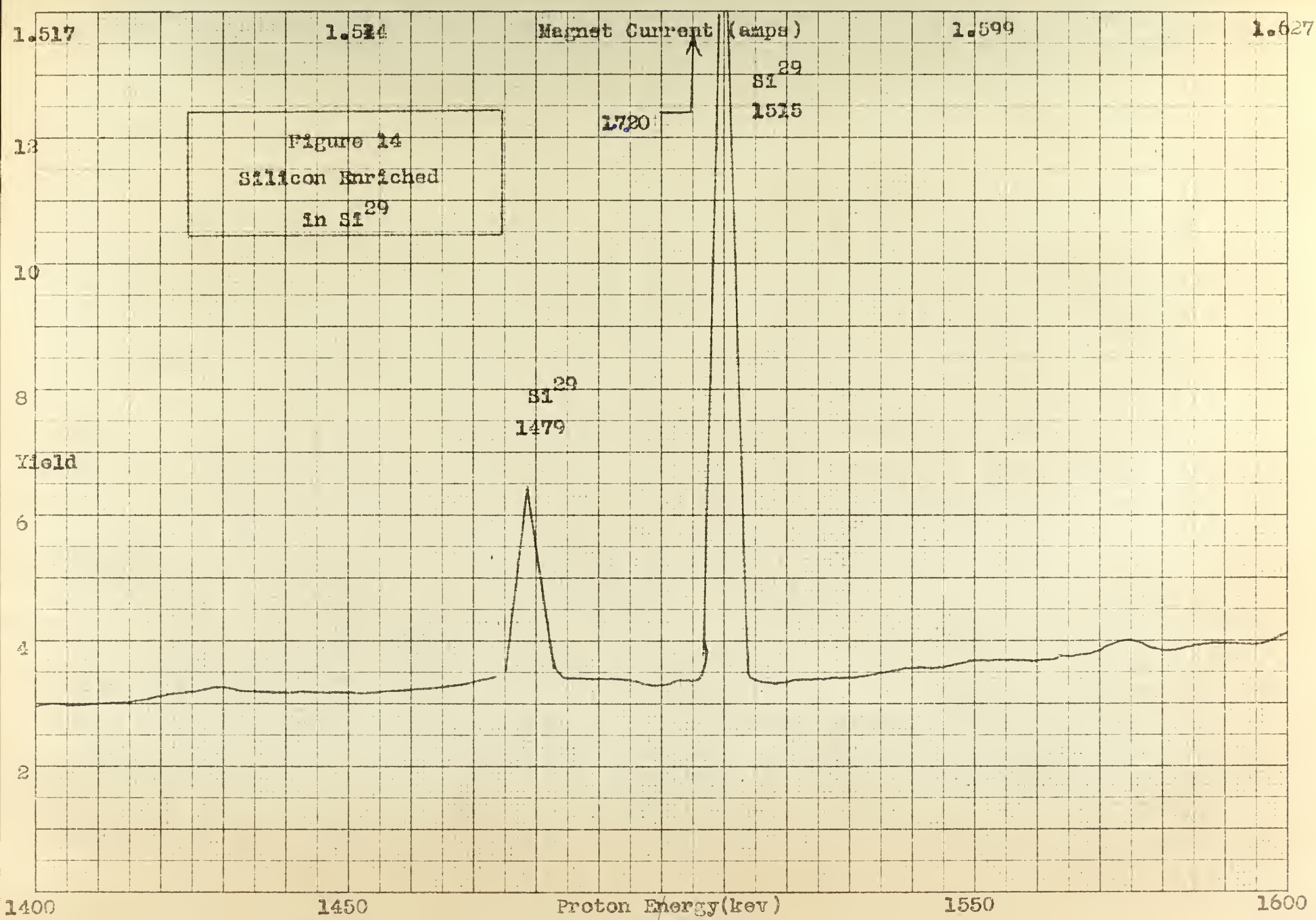


Figure 14
Silicon Enriched
in Si^{29}

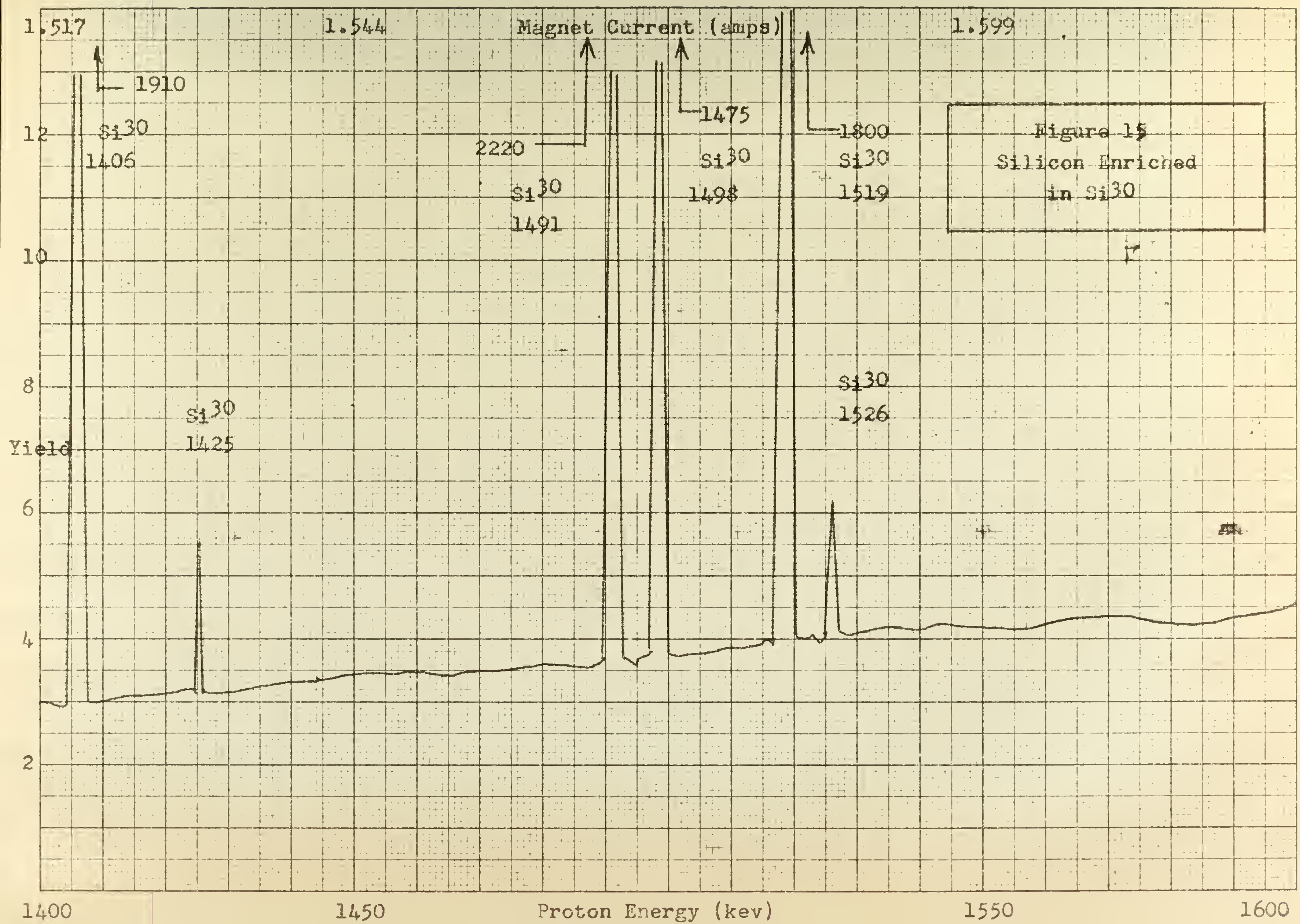


Figure 15
Silicon Enriched
in Si30
Magnet Current (amps)
1.517
1.544
1.599
1910
Si30
1406
Si30
1425
2220
Si30
1491
1475
Si30
1498
1800
Si30
1519
Si30
1526
Yield
12
10
8
6
4
2
1400
1450
1550
1600
Proton Energy (kev)

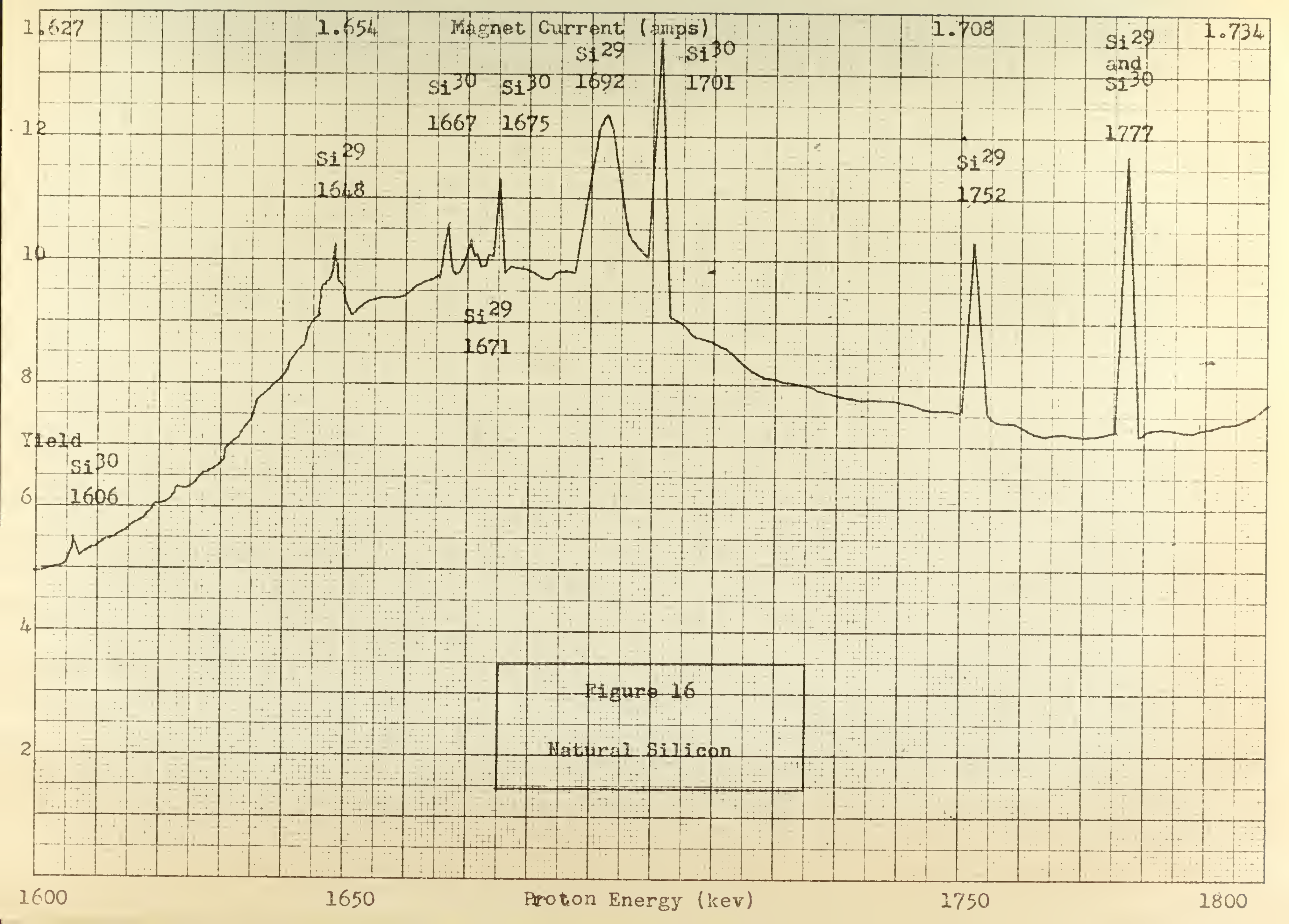


Figure 16

Natural Silicon

Figure 17
Silicon Enriched
in Si^{29}

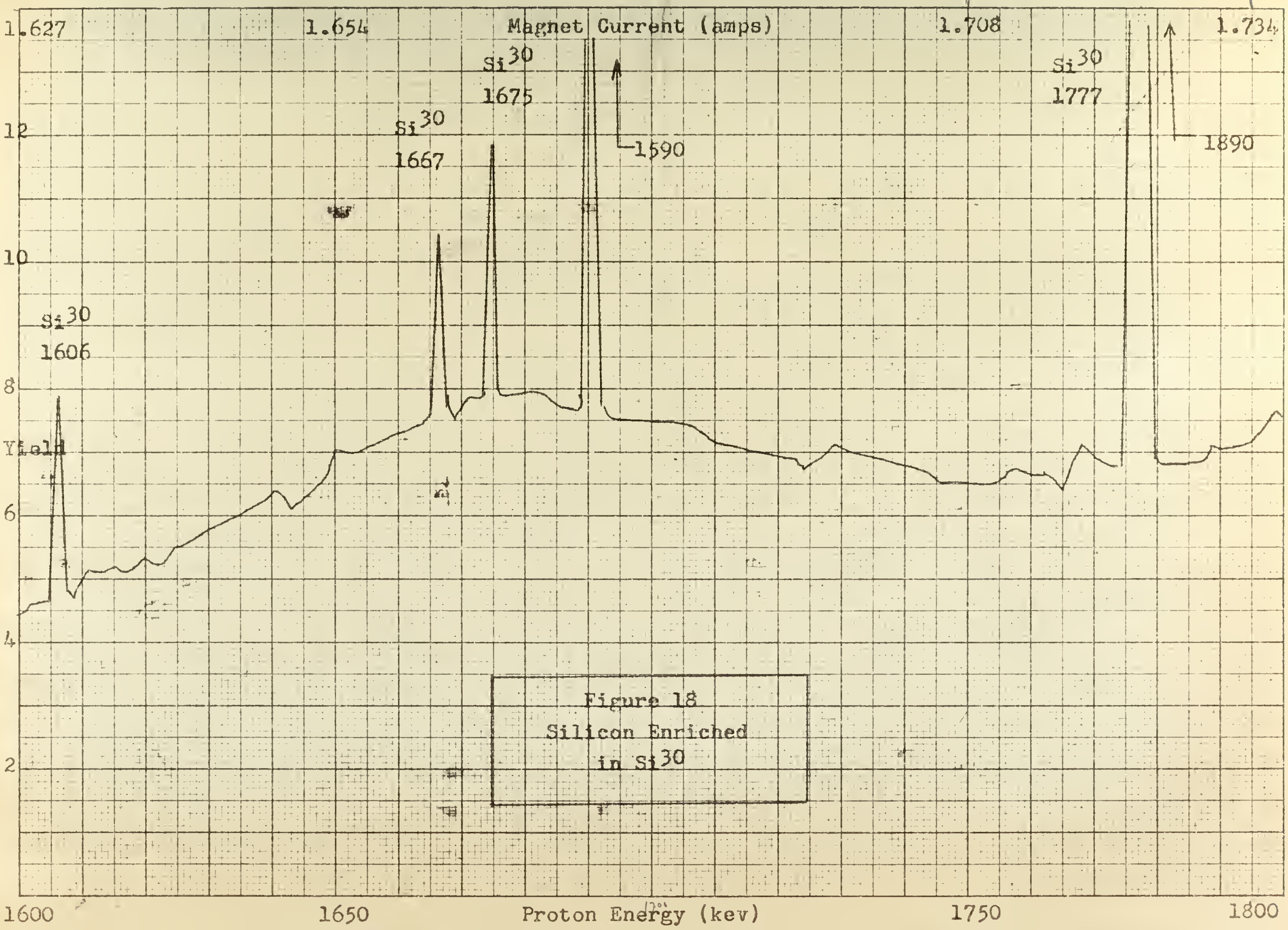


Figure 18
Silicon Enriched
in Si^{30}

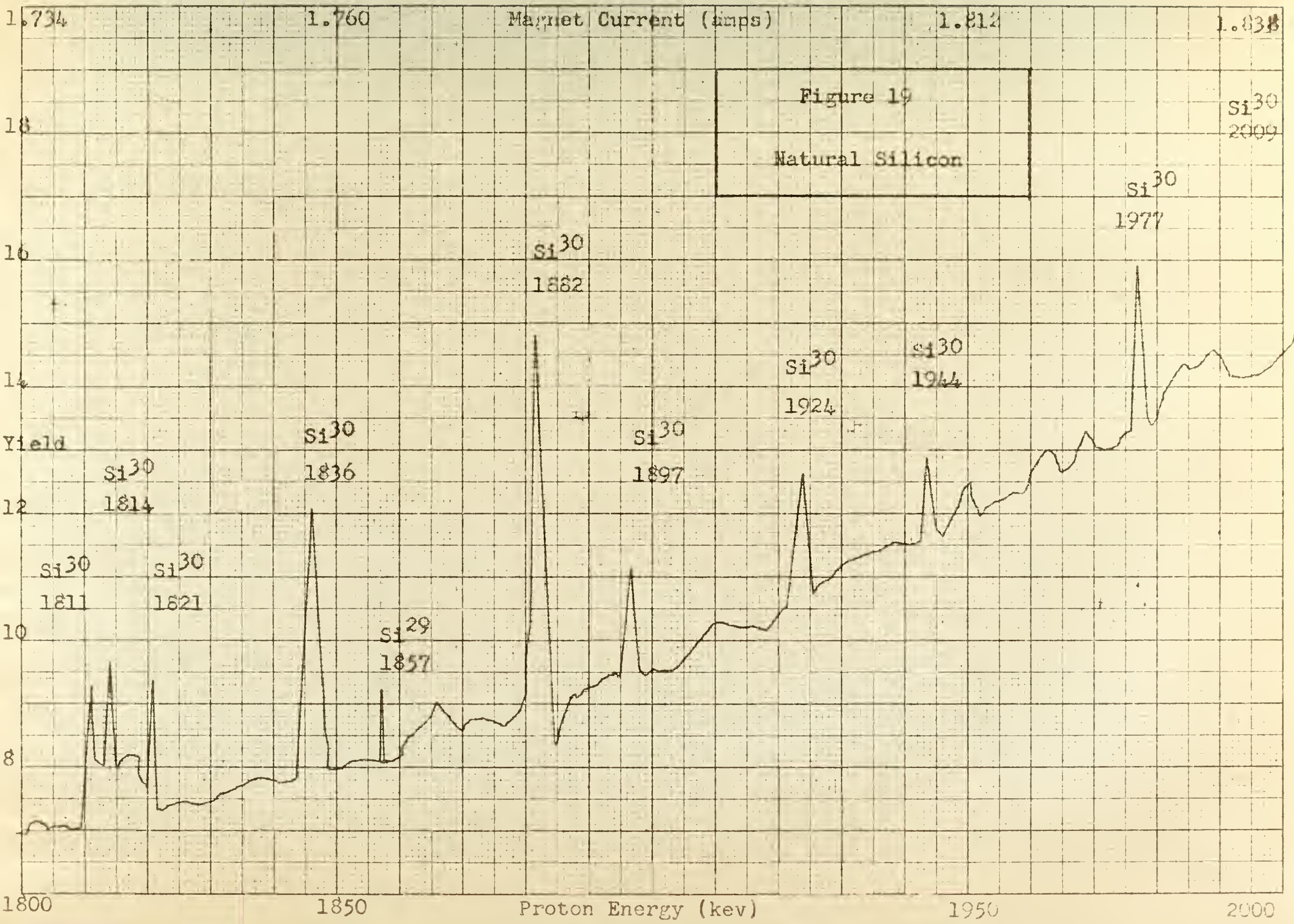


Figure 19

Natural Silicon

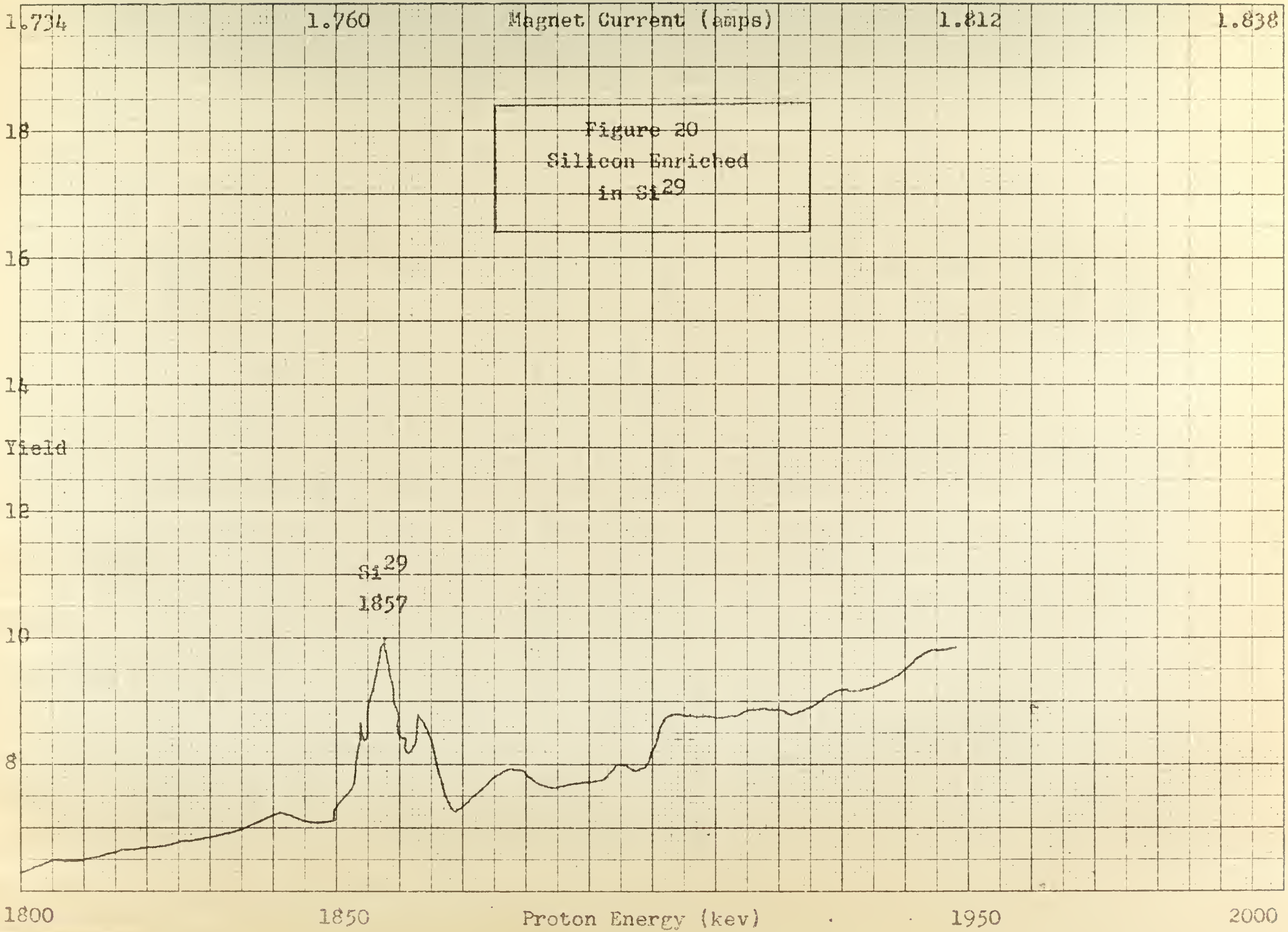


Figure 20
Silicon Enriched
in Si^{29}

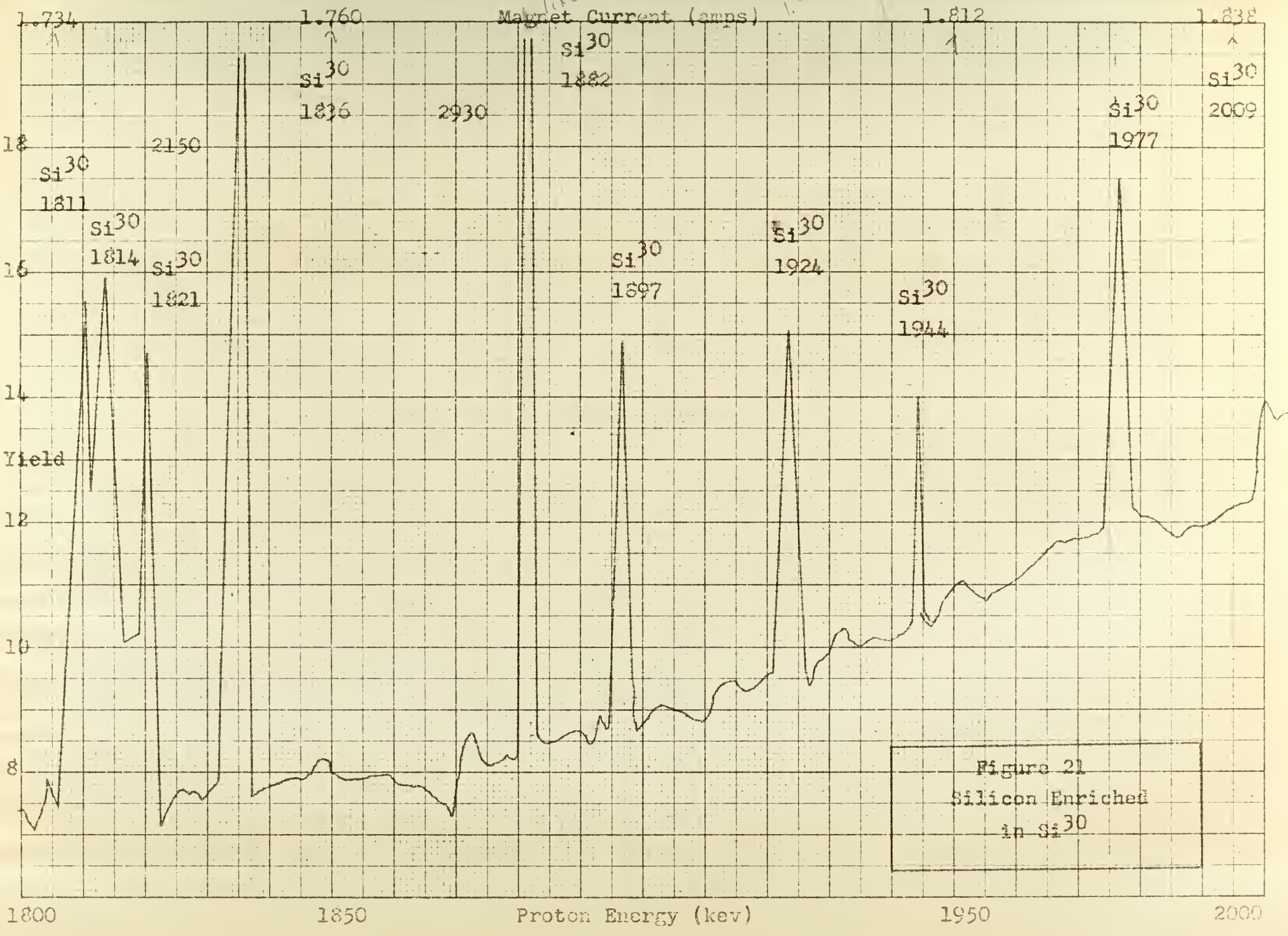
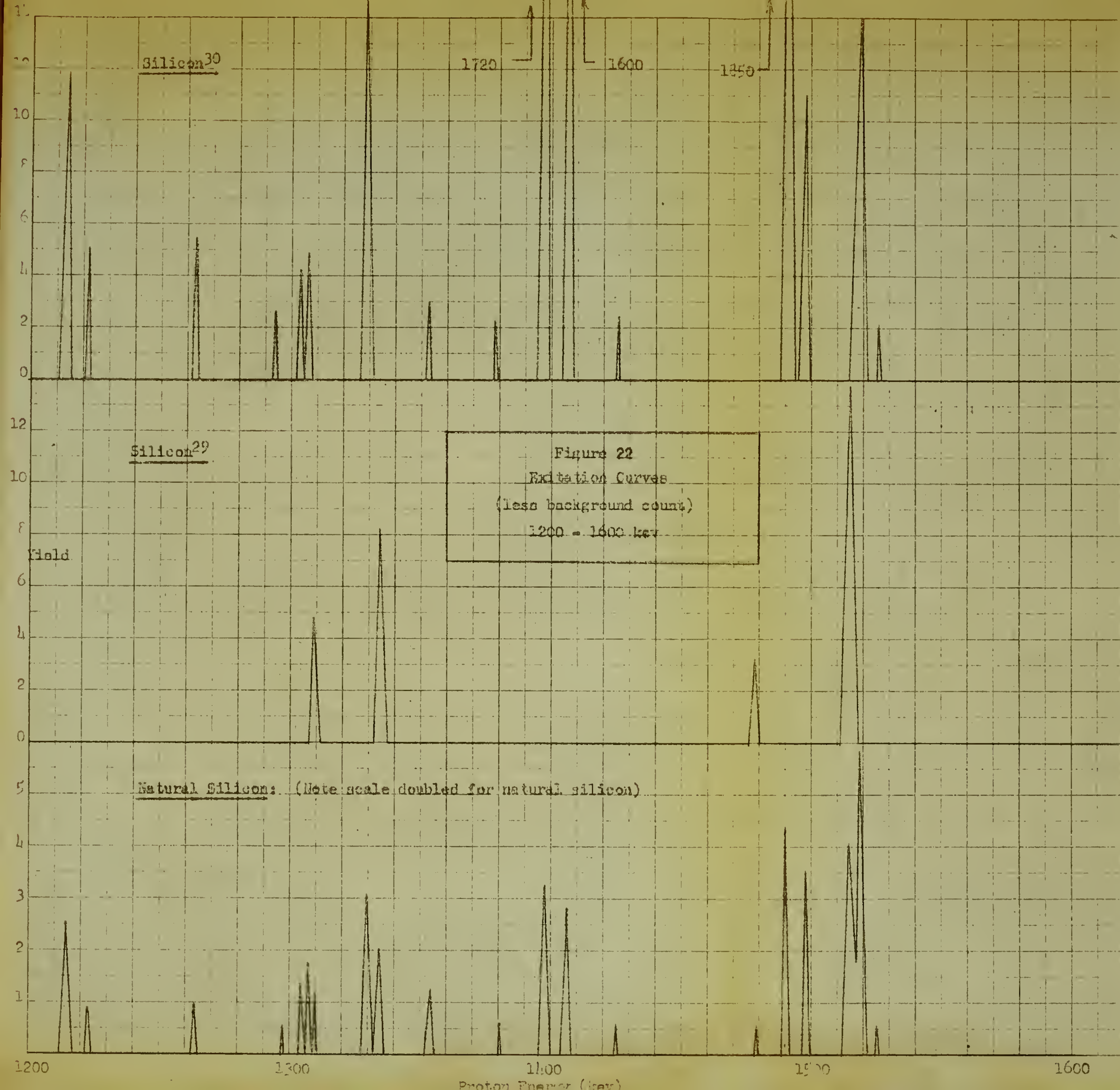
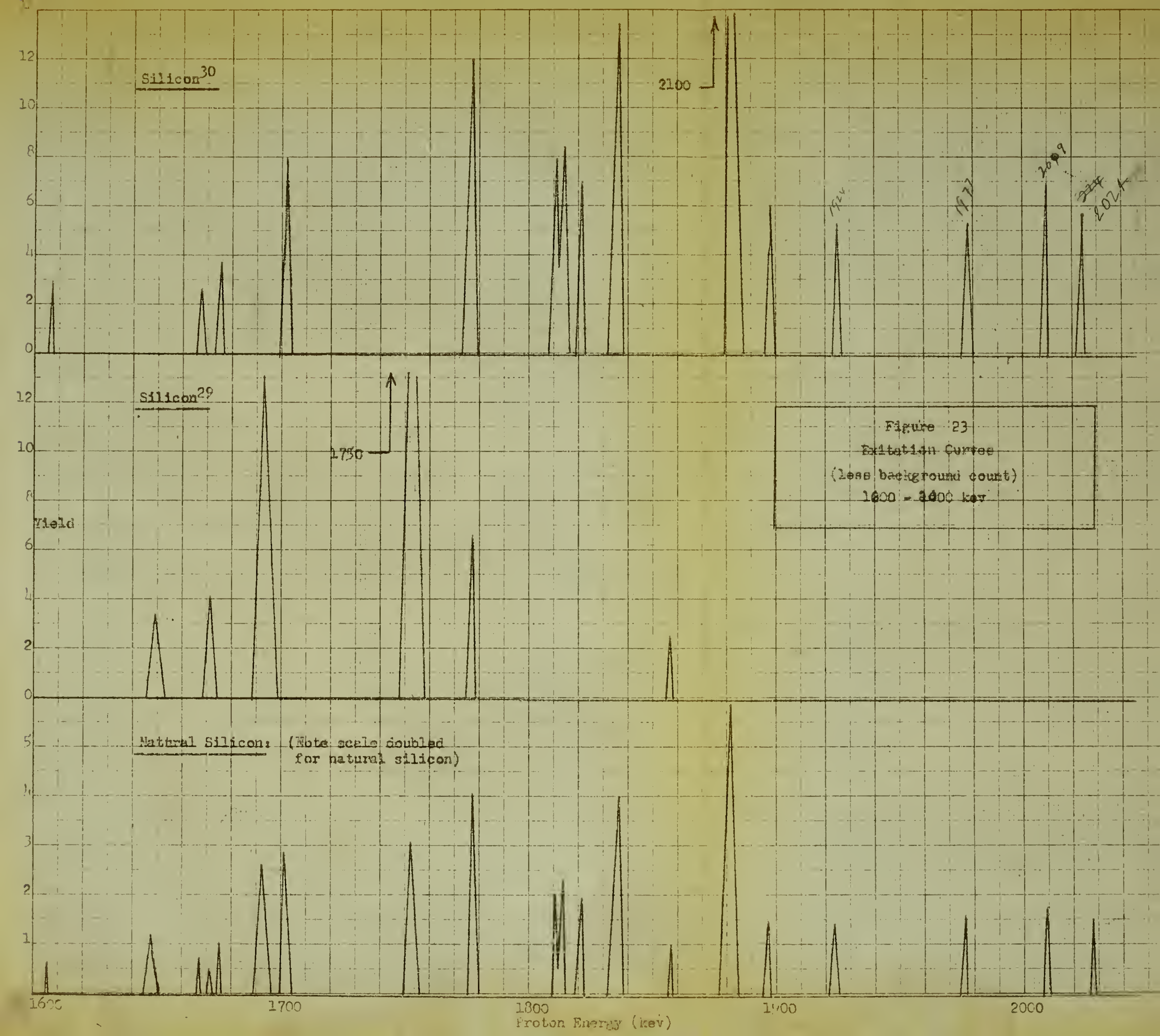


Figure 21
Silicon Enriched
in Si^{30}



Silicon30
1720
1600
1550
Silicon29
Yield
Natural Silicon: (Note scale doubled for natural silicon)
Figure 22
Excitation Curves
(less background count)
1200 - 1600 kev
1200
1300
1400
1600
Proton Energy (kev)


Silicon30
2100
Silicon29
1750
Yield
Natural Silicon: (Note scale doubled for natural silicon)
1600
1700
1800
1900
2000
Proton Energy (kev)
Figure 23
Exitation Curves
(less background count)
1600 - 2000 kev

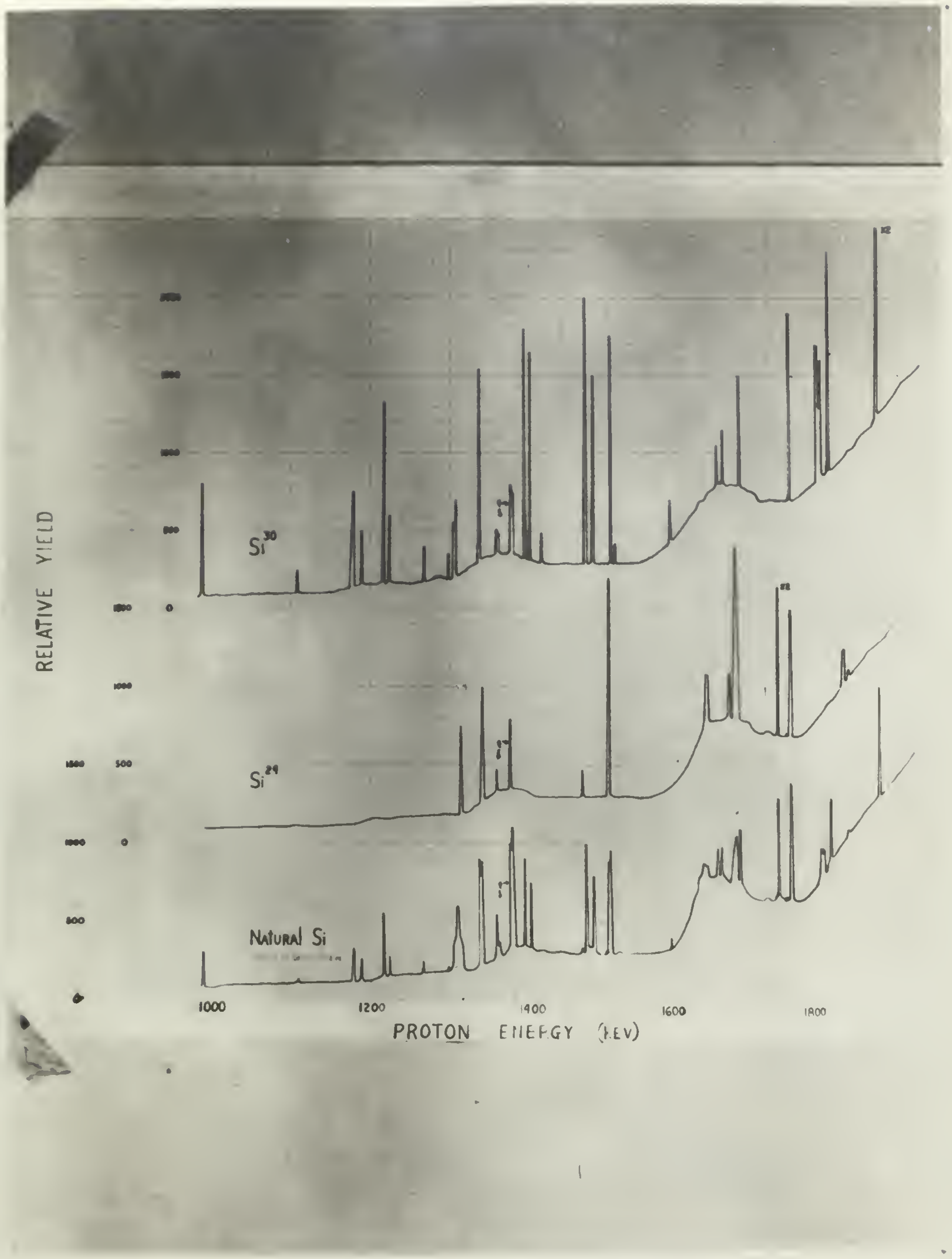


Figure 24

Photogragh of the Three Differential Exitation Curves.

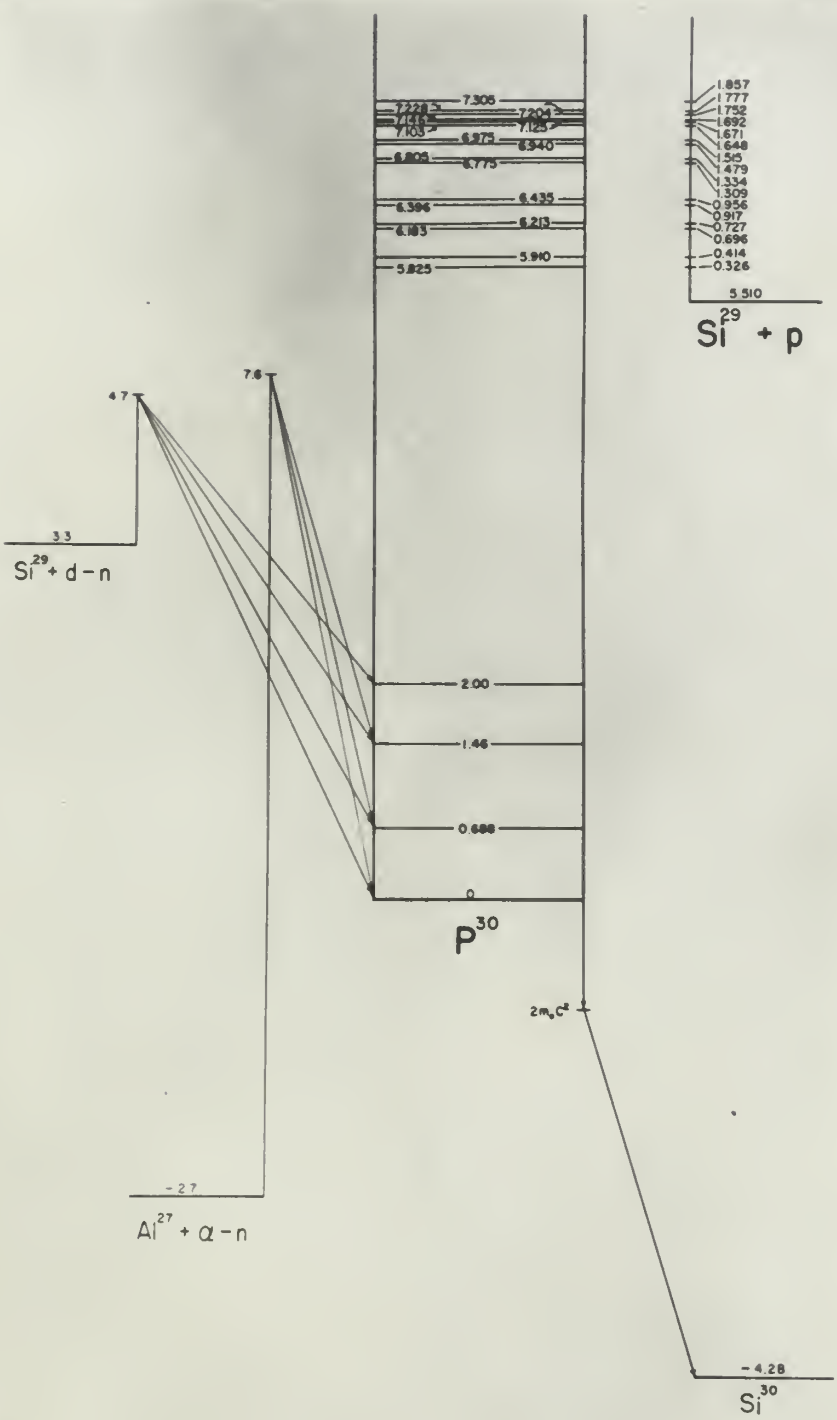


Figure 25

Photogragh of the P^{30} Energy Level Diagram

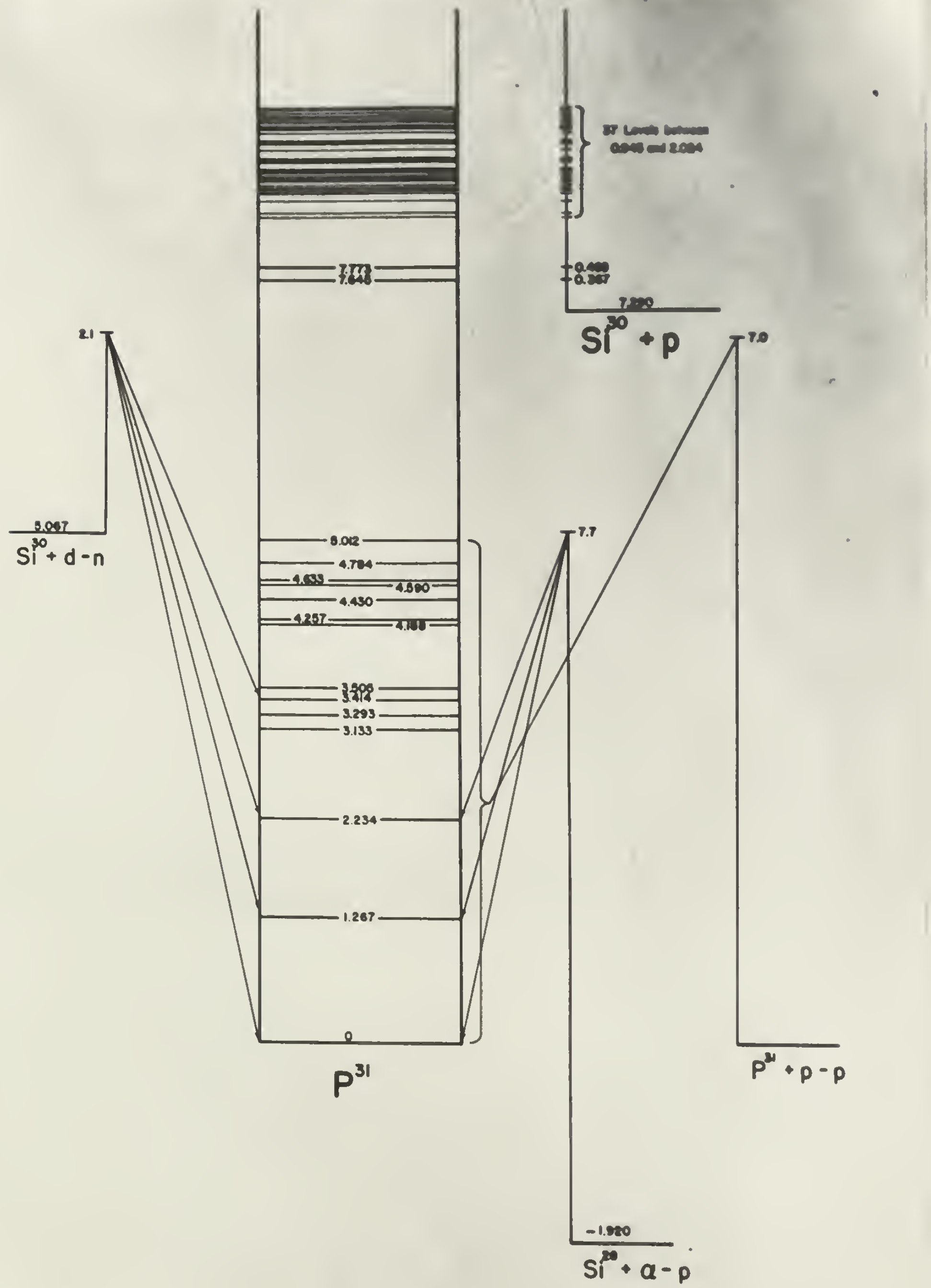


Figure 26

Photogragh of the P^{31} Energy Level Diagram